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Development and assessment of sustainable cement- and geopolymer composites

Abstract

The very energy-intensive building sector requires a substantial improvement to reduce its contribution to carbon dioxide emissions, consumption of virgin materials resources, and generation of waste materials. This principle goal still remains unsolved and only minor improvements have been attained on a global scale. A predominant paradigm based on the exploitation of natural resources together with limited assessment tools fails to make significant progress towards sustainable development. Such a challenging task requires questioning the methods of evaluation of building materials and the process of the design focused mainly on the maximization of short-term benefits. Besides that, the successful overcoming of current issues in the construction industry lies in a broader involvement of selected scientific disciplines that should participate in the process. Within this habilitation thesis, the basic principles available in the literature are summarized and commented. In the following part of the work, the contribution to the mentioned issues is described using the analysis of the replacement of cement with more eco-efficient alternatives, the potential of alkali-activated materials, and the utilization of waste alkalis as an eco-efficient alternative to commercial alkalis. Besides the development of new building materials and characterization, a combined assessment method through a combination of functional and environmental parameters is described together with the assessment of reuse scenarios for waste bricks in terms of Life Cycle Assessment. Substantial attention is devoted also to the identification of major barriers to the advances toward sustainable buildings.

Keywords: sustainability, environmental engineering, cement alternative, building sector, Life Cycle Assessment.

Navrhování a posuzování udržitelných cementových a geopolymerních kompozitů

Anotace

Stavební sektor, který je energeticky velmi náročný, vyžaduje podstatné inovace, aby došlo ke snížení jeho podílu na emisích oxidu uhličitého, spotřebě přírodních surovinových zdrojů a množství vyprodukovaného odpadu. Tento hlavní cíl zůstává stále nevyřešen a v celosvětovém měřítku bylo dosaženo pouze drobných zlepšení. Převládající paradigma založené na využívání přírodních zdrojů spolu s omezenými nástroji hodnocení nedosahuje významného pokroku směrem k udržitelnému rozvoji. Takto náročný úkol vyžaduje zpochybnit způsob hodnocení stavebních materiálů, proces jejich navrhování zaměřený především na krátkodobou maximalizaci přínosů, ale také vědní obory, které by se na procesu měly podílet. V této habilitační práci jsou uvedeny základní principy dostupné v literatuře. V následujících částech je přínos k jednotlivým problémům nastíněn z hlediska náhrady cementu alternativami šetrnými k životnímu prostředí, potenciálu alkalicky aktivovaných pojiv, využití odpadních alkálií jako ekologické alternativy komerčních alkálií, zavedení kombinované metody hodnocení prostřednictvím kombinace funkčních a environmentálních parametrů, posouzení scénářů opětovného použití odpadních cihel z hlediska posouzení životního cyklu a identifikace hlavních překážek bránících pokroku směrem k udržitelnému stavebnictví.

Klíčová slova: trvale udržitelný rozvoj, ekologické inženýrství, náhrada cementu, stavební sektor, Life Cycle Assessment

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1 Introduction

Major challenges related to present human society are strongly associated with threats caused by negative externalities of human activities such as exploitation of natural resources, adverse effects induced by pollution of the natural environment, and extensive emissions of carbon dioxide or other greenhouse gases (Allwood et al., 2010). This problem significantly affects the construction industry, as it consumes a significant share of primary energy due to the production of energy-intensive binders, high demands on indoor temperature comfort, etc. (Finch et al., 2021). Among others, a huge virgin material demand harms the natural environment in terms of natural resource depletion and creates a burden on future generations (Pome et al., 2021). Specifically, approximately € 240 billion in damages were caused by different and indirect side-effects of human activities on planet Earth (Biskaborn et al., 2019; Ricke et al., 2018).

To reflect the above-mentioned tasks sufficiently, the sustainability principles have been developed and adopted in several fields including the construction sector, the materials design in particular. In fact, a more responsible approach regarding the preservation of the natural environment represents a major research effort during the last decades in the field of materials engineering (Bahramian and Yetilmezsoy, 2020). Within this paradigm change, new approaches have been established and environmental factors have been adopted for modern building materials design (Passoni et al., 2021). Considering principles of sustainable development, the solution strategy of this problem was gradually extended to additional scientific disciplines related to sustainability pillars as provided in Figure 1. In this sense, the interconnection between particular pillars was mentioned to reveal and quantify the results of indirect effects or so-called positive and negative externalities of human activities (Cabo et al., 2020). For example, extensive exploitation of natural resources is accompanied by adverse effects on human health (cement production induces an increased cancer occurrence), therefore it may result in increased costs spent on healthcare, reduced labor productivity, the necessity of landscape restoration, and many other issues (Rentschler and Bazilian, 2017). In other words, issues associated with indirect costs/benefits were not taken into account sufficiently.

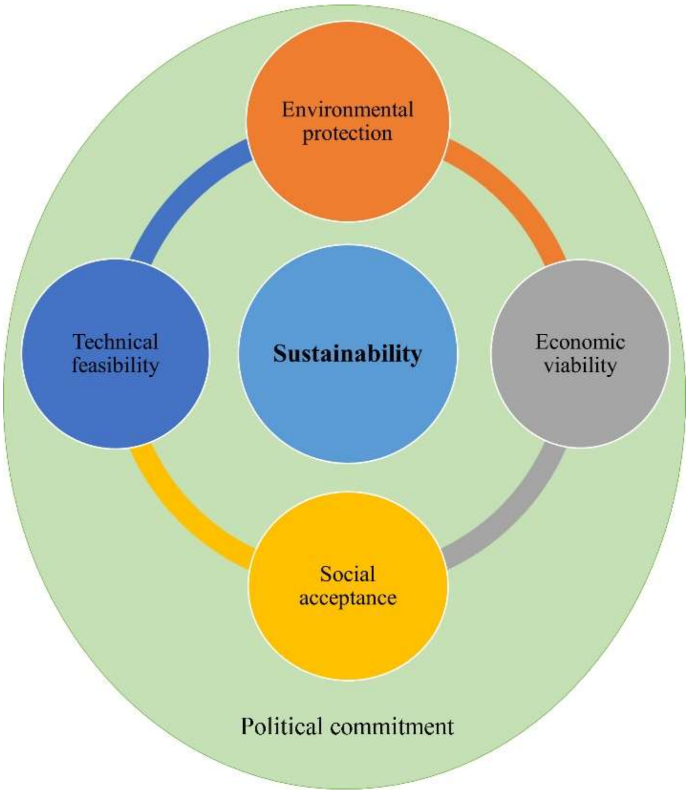

Figure 1. Basic pillars of sustainability (Fort and Cerny, 2022)

Besides problems related to rational and effective utilization of natural resources, the fast rate of generation and accumulation of produced waste can be viewed as a substantial threat to future generations (Fort and Cerny, 2020; Foster et al., 2021; Kumara et al., 2018). In this regard, the responsible design taking into account whole service life (production, use, disposal at end-of-life) should be applied to a greater extent.

Building operation optimization has attracted substantial attention within the last decades in order to improve the thermal resistance of building envelopes (Finch et al., 2021; Perez-Bella et al., 2020). This effort goes hand in hand with increased requirements of building occupants on indoor thermal comfort and thus a high energy consumption spent on building heating and cooling. In this sense, the issues associated with the preservation of indoor comfort resulted in the development of a wide range of highly efficient insulation materials (Abu-Jdayil et al., 2019; Asdrubali et al., 2019). In other words, the application of thermal insulation material was found as a viable scenario leading to significantly reduced energy consumption. Together with insulation materials, several advanced materials have been investigated to improve thermal inertia through the utilization of phase change materials (PCMs) (Rathore et al., 2020; Sharma and Rai, 2020). While the thermal insulation materials act rather as passive elements in the building envelope, the application of PCMs is capable to absorb/release a certain amount of energy during temperature peaks and contributing to the maintenance of ambient conditions. It should be noted that new buildings must comply with demanding energy-efficiency standards and therefore contribute to the reduction of the energy intensity of the building sector and improvements can be done rather by reconstruction of the outdated buildings (Huo et al., 2021).

Viewing the advances in the operating efficiency of the modern building in terms of heating/cooling demands (as the most contribution process to energy intensity) and superior thermal insulation properties of modern materials, substantial advances need to be done rather in the production of materials and the disposal scenarios.

2 Sustainable concrete and geopolymer production: a short survey of recent studies

The construction materials production is extremely dependent on the availability of natural resources, thus significant advances to leave this paradigm and transform it into a more sustainable circular economy model are required. Accordingly, the replacement of virgin materials represents a very important challenge to reduce the environmental impact (Jalilifar and Sajedi, 2021; Shah et al., 2021). The utilization of various by- or waste products has been found as an efficient way toward lowered virgin materials exploitation and at the same time, a reduction of waste production. In these terms, two basic approaches can be distinguished; replacement of natural aggregates; and partial replacement of cement by alternative materials or completely avoided cement production by alkali-activation (Zamora-Castro et al., 2021).

2.1 Replacement of natural aggregates

Natural aggregate replacement was considered the first step due to the high volume required for concrete production. Moreover, low requirements on its quality (compared to cement replacement) improve the attractivity for the real application. Across the literature is possible to distinguish many research papers contemplating the natural aggregates replacement, and related environmental benefits accessed through the Life Cycle Assessment (LCA) and Life cycle Cost (LCC) analysis (Di Maria et al., 2018; Meng et al., 2018; Wong et al., 2018). Notwithstanding, issues related to strength, elastic modulus, toughness, stress-strain relationship, water absorption, etc. of materials with alternative substituents with natural aggregates represent factors that limit the utilization of recycled aggregates (RCA) in concrete. Due to the attractivity of this topic, many publications describe the effects of various types of RCA on the mechanical performance, formation of the interfacial transition zone between RCA and cement matrix, water transport properties, the thermal conductivity of cement-based composites as well as the reduction of environmental impact can be found in the literature (Bouarroudj et al., 2021; Jalilifar and Sajedi, 2021; Kirthika and Singh, 2020; Meddah et al., 2020; Mikhailenko et al., 2020). The list of widely studied RCA alternatives is provided in Table 1 along with replacement ratio and effects on mechanical strength. The provided list does not include a complete list of natural aggregate replacement alternatives and many others can be distinguished including waste plastic, rubber, and various agricultural products.

Table 1. Natural aggregate alternatives

Material	**Density**	**Used Replacement**	**Compressive strength of composite**	**Reference**
Expanded perlite (EP)	30 to 150 kg/m^3	0 to 20 % EP added to OPC containing SCM (50 % slag + 7 % silica fume).	From 23.69 MPa to 41.58 MPa	Ibrahim et al. (2020)
		30, 40, 50 % EP replaced river sand aggregate.	54.2 MPa – 50 % replacement 31.8 MPa - 30% replacement.	Mathawee et al. (2013)
		0 to 80 % EP replaced sand aggregate.	18.5 MPa – 15% replacement.	Jedidi et al. (2015)
Pumice	500 to 600 kg/m^3	Replacement of coarse aggregate by 50% pumice.	15 MPa	Top et al. (2018)
		Replacement of high calcium fly	2.7 to 7 MPa	Wonga et al. (2018)

		ash containing crush clay brick.		
		Replacement s; > Lightweight coarse and natural fine sand aggregate (LCNF). > lightweight coarse and fine aggregate concrete(LCF)	LCNF (20.4 to 30.2 MPa) LCF (15.9 to 19.6 MPa) Control 36 MPa	Parhizkar et al. (2012)
Diatomite	500 to 928 kg/m^3	Clay brick replaced		Hasan et al. (2021)
		Added to OPC	7.8 to 12.9 MPa	Posi et al. (2013)
		OPC with SCM (fly ash and silica fume) was substituted.	51 MPa	Tagnit- Hamou et al. (2003)
Exfoliated vermiculite	64 to 160 kg/m^3	Exfoliated vermiculate substituted Sintered fly ash.	20.3 MPa to 54.2 MPa	Przychodzien and Katzer (2021)

		Light weight cement based mortars.	3.9 MPa and 16.4 MPa	Koksal et al. (2015)
		The natural river sand fine aggregate replacement.	14.80 MPa	Schackow et al. (2014)
Expanded clay	250 kg/m^3 to 510 kg/m^3	Sand aggregate in fly ash base concrete replaced	55 MPa to 60 MPa	Li et al. (2012)
		Added to self-compacting lightweight concrete,	37 – 61 MPa	Bogas et al. (2012).
		partially and fully replacement of coarse aggregate by expanded clay 20 to 100%	28.56 MPa - 40% 21.77 MPa - 100% replacement	Ashokkumar et al. (2021)
Coal bottom ash/boiler slag	641.03 to 1440 kg/m^3	Added to fly ash geopolymer concrete.	14.3–18.1 MPa	Wongsa et al. (2016)
		0 to 40% Partial replacement of	49.5 MPa - 10% 26 MPa - 40% replacement	Ghazali et al. (2022)

		river sand at 60 days of curing.		
Expanded polystyrene (EPS)	11 to 34 kg/m^3	Added to mortars containing silica fume (SCM).	10 to 21 MPa	Babu and Babu. (2003)
		EPS added to OPC mortar.	15.55MPa	Schackow et al. (2014)
Sintered fly ash	645 to 755 kg/m^3	Sand aggregate replaced.	20.3 MPa to 54.2 MPa	Przychodzien and Katzer (2021)
		Fine aggregate replaced with sintered fly ash sand.	Specific gravity: 1.57 g/cm^3	Ojha et al. (2021)
		Replaced broken granite-crushed stone as coarse aggregate to OPC.	36.25 MPa - 12% addition. 26.68 MPa - 20%	Kumar and Kumar (2014)
Blast furnace slag	800 to 1040 kg/m^3	Added to metakaolin-fly ash binders in ratio 1:1.	4 MPa - 28 days	Aguilar et al. (2010)

		Replaced coarse sand with furnace slag aggregate	41 MPa – 28 days of curing.	Lofty et al. (2015)
Glass broken/cullet	600 to 1330 kg/m^3	Fine sand aggregates, cement and both were replaced.	35 MPa	Al-Kizwini (2020)
		Partial replacement of fine aggregate	32.94 MPa - 10% replacement	Rahim et al. (2014)
		Partial replacement of fine aggregate (10,20, 30 %)	32.85 MPa - 30% replacement	Mishra et al. (2020)

Considering the environmental impact, available information based on commonly used eco-indicators exhibited substantial improvements if transport distance up to 60 km is considered. In other words, the LCA result outputs are highly sensitive to transportation distance, and employment of any advanced and energy-consuming machinery (Yazdanbakhsh et al., 2018). Notwithstanding, downcycling of end-of-life materials with high embodied energy (for example concrete or bricks) to RCA used for low-grade road application was found by Di Maria et al. (2018) as a scenario with very limited environmental benefits (given mainly by avoided production of natural aggregates). The authors noted that the limited availability of a high-quality RCA prevents more advanced recycling options and destinations for such materials. On the other hand, sustainable management needs to be aimed at more advanced recycling options preferring recycling instead of downcycling with questionable future prosperity.

2.2 Replacement of a part of Portland cement by SCM

A principal condition for solving the current problem with a high energy intensity of binders' production can be seen in the availability of technological solutions and a robust knowledge base. Therefore, a significant effort was spent on the identification of available materials that can be utilized as a traditional binder replacement, Portland cement in particular (Bostanci, 2020; Nwankwo et al., 2020). A significant part of scientific research in the field of materials engineering is aimed at the utilization of various by-products and waste materials as cement replacement (Portland cement production represents about 8 % of global carbon dioxide emissions) to deliver sufficient cost and environmental benefits. Taking into account the fact that the Portland cement production already exceeded the annual production of four billion tons, the cement replacement represents a truly challenging issue (Hamada et al., 2018). Moreover, besides savings related to cement replacement, the utilization of industrial waste or by-products improves the overall recycling rate and thus reduces landfilling. Among the most investigated alternative materials, often called supplementary cementitious materials (SCMs), coal fly ash, metakaolin, silica fume, ground granulated blast furnace slag, ceramic brick waste, municipal waste ash, rice husk ash, palm oil fly ash, and several other alternatives can be recognized (Hansen and Sadeghian, 2020; Lee et al., 2020; Liu et al., 2020; Palod et al., 2019). As reported in the literature, various effects are assigned with the use of SCMs in concrete. On the one hand, improved long-term strength, reduced permeability, and lower probability of alkali-silica reaction can be expected in such modified composites. On the contrary, the worsening of rheologic properties, undesired changes in the initial and final setting time, ettringite formation, and degradation in the strength parameters have been recognized (Lothenbach et al., 2011; Mo et al., 2017). In general, the SCMs can be divided into two categories, self-cementing materials which react with water similarly to Portland cement, and primarily silicon-based pozzolans which require besides the water also the presence of calcium. The chemical composition of selected SCMs is provided in Table 2.

Table 2. Chemical composition of pozzolan materials

Material	**Content (%)**								**Reference**
	SiO_2	**Al_2O3**	**Fe_2O_3**	**CaO**	**MgO**	**SO_3**	**Na_2O**	**K_2O**	
Metakaolin	51.52	44.53	0.48	0.02	0.19	–	0.29	0.16	Villaquiran-Caicedo (2019)
Diatomite	80.3	6.1	6.79	1.04	0.65	0.12	0.62	0.44	Hassan et al. (2019)
Fly ash	55.38	28.14	3.31	3.45	1.85	0.32	2.3	1.39	Gholampour et al. (2019b)
Ground granulated blast furnace slag	18.9	6.43	0.74	66.9	1.41	1.97	–	0.67	Alrefaei et al. (2019)
Red mud	16.51	28.05	30.32	2.22	0.7	–	8.7	0.26	Yang et al. (2020)
Rice husk ash	92.33	0.18	0.17	0.63	0.49	–	0.07	0.15	Villaquiran-Caicedo (2019)
Olive biomass fly ash	12.6	2.97	2.3	20.21	4.85	4	1.12	26.34	Alonso et al. (2019)
Volcanic ash	43	15	12	11	6.8	–	4.6	1.7	Lemougna et al. (2020)
Silica fume	90	1.2	2	1	0.6	0.5	–	–	Cheah et al. (2019)

Sewage sludge ash	38.28	20.72	11.27	5.51	1.91	4.18	0.7	0.73	Istuque et al. (2019)
Steel slag	11.9	1.23	31.85	45.72	3.16	0.22	0.03	0.03	Chen et al. (2019)
Wood biomass ash	21.86	7.29	2.56	29.36	2.55	4.76	2.54	3.89	Hassan et al. (2019)
Palm oil fly ash	64.2	4.25	3.13	10.2	5.9	0.09	0.1	8.64	Huseien et al. (2016)
Glass powder	3.19	1.24	0.24	63.35	0.16	0.64	0.28	0.01	(Khan et al., 2020)
Red brick powder	60.31	15.61	7.72	5.6	3.05	–	0.56	4.48	(Tang et al., 2020)

While the hydration of Portland cement is described in detail and consists of 4 main stages (initial rapid dissolving, induction period, acceleration period, post-acceleration period), and formation of C-S-H gel, the added pozzolans react with calcium hydroxide and form calcium aluminate hydrate next to calcium silicate hydrate (Ramanathan et al., 2020). Such modification of the hydration process can be linked with notable benefits if appropriately applied to the concrete. Several SCMs are accompanied also by the lowered hydration heat produced by tricalcium silicate hydration, therefore mitigating the crack formation due to the thermal gradient between the inner structure and exterior (Shakouri et al., 2020). To access the material suitability for pozzolanic reaction, high silica and alumina content were understood as the most crucial parameters influencing the pozzolan activity. Notwithstanding, recent works refer also to the particular importance of the amorphous content, as the crystalline silica part cannot be deemed as soluble in cement pore solution (Zafar et al., 2020). As revealed, the oxide composition should be always completed by the Rietveld analysis of X-ray diffraction data that allows the identification of the phase composition (Keppert et al., 2018). Besides this method, Snellings et al. (2017) introduced an innovative method based on differences between the unique 2θ values of amorphous and crystalline phases. As the determination of the pozzolanic activity represents the rule of the thumb, several methods have been developed including the measurement of the consumption of the portlandite in SCM- cement paste, reactivity in lime solution (Chapelle test), reactivity in cement-water solution (Frattini test), the combination of the previous two methods, determination of the activation energy, and so on. In terms of improvement, various methods such as thermal activation, additives application, and particle size reduction have been studied over the last decades to improve the material reactivity and attractiveness as cement replacement alternatives. The suitability of selected SCMs for cement replacement in building composites is assessed in Table 3.

Table 3. Effects of cement replacement using selected SCMs

SCM	**Replacement level**	**Results**	**Reference**
Blast furnace slag	10 % - 90 %	Reduction in compressive strength was about 10 % up to 20 % replacement; further increase in replacement level up to 60 % reduces the strength by about 29%.	Mo et al. (2015)
		28-day compressive strength of concrete modified by 30 %, 50 %, 70 % and 90 % slag were 1.4%, 4.2%, 11.2% and 14.6% lower than the control concrete.	Karahan (2017)

Fly ash	5 % – 40 %	Results indicate a minor reduction of the compressive strength values even after 40% replacement. Replacement of Portland cement with original fly ash increased the porosity but decreased the average pore size of the paste.	Chindaprasirt et al. (2005)
Rice husk ash	10 % - 30 %	Optimum compressive strength (increase of up to 9%) was found at 10% replacement.	Nasiru et al. (2021)
		Replacement level of 30% exhibited comparable strength with control specimens; further replacement decreased the compressive strength.	Alex et al. (2016)
Metakaolin	10 % - 30 %	Metakaolin significantly increased CSH content up to a replacement level of 20 %, while replacement levels over 30 % appeared to reduce CSH formation.	Mikhailenko et al. (2018)
Rice husk ash	10 % - 30 %	cement paste with metakaolin content achieved comparable 28-day strength to that of the reference even at 40 wt.% replacement level.	Zhao and Khoshnazar (2020)
Sewage sludge ash	10 % - 25 %	Replacement of 10 % cement by sewage sludge ash resulted in a 25% improvement in compressive strength compared to control samples.	Swierczek et al. (2021)

Waste glass powder	5 % - 25 %	Insignificant effect on setting time and cement expansion Mortars with replacement up to 10 % increased compressive strength about 9 %.	Aliabdo et al. (2016)
Sewage sludge ash	10 % - 30%	In the fresh state, the partial replacement (10 and 20%) of Portland cement by glass waste had no significant impact.	Jochem et al. (2021)
Wood biomass ash	10 % - 50 %	Beyond 30% in replacement, mechanical and physical properties decreased at a significantly higher rate.	Vu et al. (2019)
		Replacement of 40% wt. of cement resulted in a lower compressive strength, but still compliant with the requirement of 32.5 MPa	Tosti et al. (2018)
Wood biomass ash	10 % - 50 %	Flexural strength of samples with 15 % - 25 % at 365 days exhibit lower values compared to control sample. Samples with 5 and 10 % achieved comparable results with control sample.	Ruiz-Sanchez et al. (2019)
Waste marble ash	5 % - 25 %	Compressive strength of mixes with 5 % and 10 % was increased about 7 %, 12 % respectively. Compression resistance at	Aydin and Arel (2019)

		curing ages of 90 and 365 days was >50% of reference resistance values at 28 days.	
Diatomite	5 % - 40 %	Replacement up to 40 % did not affect compressive strength, tensile strength was improved.	Ahmadi et al. (2018)
Red mud	0 – 10 %	Negative effects on fresh properties, about 19% improvement in compressive strength.	Ghalehnovi et al. (2019)
Waste brick	10% - 50%.	Similar compressive strength as the control cement paste was observed for cement paste with up to 20% brick dust.	Lin et al. (2010)
		Mortars with replacement up to 10 % have comparable 90-day compressive and flexural strengths as the control mortar.	Yang et al. (2020)
		Increase from 10 % to 30 % of brick dust reduced about 25 % compressive strength of mortar.	Tang et al. (2020)

To highlight the major effects associated with SCMs application, several findings can be drawn. First, the pozzolanic reaction of the various SCMs revealed lower hydration kinetics compared to Portland cement, however sufficient grinding fineness can increase the material pozzolanic activity and the specific surface area (Komnitsas et al., 2015; Vejmelkova et al., 2012; Čáchová et al., 2014). The workability of concrete modified by SCMs goes along with detrimental effects and thus some limitations for the real application. The increased dosages of water associated with silica fume, or some biomass ashes sometimes overweight the benefits, and higher dosages of superplasticizers or optimization of the particle size distribution are required to provide materials with satisfactory functional parameters. In terms of functional parameters, strength, in particular, SCMs are usually accepted as materials contributing to long-time strength improvements thanks to pozzolanic reaction and decrease. On the other hand, the dilution of cement results usually in a reduction in the early age strength according to several authors. As reported by Cantero et al. (2020), the application of dosages between 5 % and 20 % of metakaolin provides an improvement in compressive strength up to 40 % compared to only Portland cement concrete. Similarly, these effects are visible in the case of later age strength testing, when the application of blended cement with limestone and metakaolin, achieved more favorable strength parameters. Results obtained for other cement alternatives such as rice husk ash or waste brick powder in similar rations point to the potential of these materials as well (Sandhu and Siddique, 2017; Yang et al., 2021). As ensued from literature, the cement can be up to 20 wt.% successfully replaced without significant worsening of the functional parameters even some improvements can be expected. Performed analysis of formed hydrates revealed a dominant share (dependent on the amount of used SCM) of calcium silicates C-S-H also present in ordinary Portland cement mixtures, aluminates (C-A-H), and aluminosilicates (C-A-S-H) (Navratilova and Rovnanikova, 2016). Aluminates and aluminosilicates can be viewed as products of pozzolanic reactions responsible for the growth of mechanical strength at later ages (Ge et al., 2012). In these terms, various authors used single, binary or ternary blended concrete in order to deliver satisfactory results in terms of compressive strength, modulus of elasticity, or permeability. However, it should be mentioned that the incorporation of SCMs during concrete production is associated with more complicated hydration processes. On the contrary, a higher replacement level is often accompanied by more pronounced changes in the material microstructure as well as the deterioration of the functional performance. Specifically, with some variations, a replacement level up to 30 wt. % can be accepted as a threshold value and higher cement replacement goes along with a rapid reduction in strength parameters, a shift in material porosity, permeability, etc. (He et al., 2020; Piemonti et al., 2021).

Besides the functional parameter's modification, sufficient attention should be paid to the significantly lowered carbon dioxide footprint as well as cost savings as the original motivation of the cement replacement. Considering the environmental or financial cost of SCMs, most of them are accepted as cost-free excluding transportation (Caldas et al., 2021). Therefore, the replacement of the Portland cement content in concrete up to 30 wt.% (in the very positive case) provides potential benefits in terms of energy savings or CO_2 emissions to the same extent (neglecting the possible treatment of SCMs and the material transport). Although this fact, the geographic availability of particular SCMs should be considered due to local specifics of the industry, biomass production, and other regional factors.

As accessed in recently published studies, the carbon dioxide emissions savings are the most investigated parameters describing the correlation between effects on the natural environment and the Portland cement replacement. Specifically, Alnahhal et al. (2018) revealed saving of about 12 – 14 % of palm oil fly ash or rice husk ash is used as the cement replacement, and drying, grinding, and transportation processes are included in the analysis. The importance of the calculation of transportation was emphasized by McLellan et al. (2011) however, since the concrete industry represents a well-established industry in terms of the short bulk material transport, efficient machinery, and supply/demand chains, the direct comparison of both products does not include the maturity of products (development of SCMs based cement is accompanied with the rationalization of the materials flow, improvement in used machinery, fuel efficiency, etc.). Similarly to CO_2 saving, LCA analysis was employed by Panesar et al. (2019) to highlight the benefits of the utilization of coal combustion fly ash up to 50 wt. %. As the LCA analysis delivers more robust results in terms of environmental burden, the impact categories as Ecotoxicity, Human toxicity, and Resources and fossils refer to linearly increased results in line with applied replacement level as well as the transportation distances. Apparently, the most favorable results are linked with 50% replacement.

Notwithstanding, the utilization of waste products brings issues related to the fact that the applicability of coal fly ash was questioned due to the content of hazardous compounds (heavy metals in particular) and its limited availability in the close future (Fort et al., 2021). In this regard, biomass fly ash may be of particular attention thanks to the lowered concentration of hazardous compounds including the development of biomass combustion in the EU. Notwithstanding, the deterioration of material properties associated with higher BFA dosage prevents the achievement of substantial advances in terms of mitigation of the environmental burden.

It should be noted that a vast majority of listed alternatives provide satisfactory functional performance up to 30% replacement only and cannot be deemed as a viable scenario in terms of zero-carbon targets (Huo et al., 2021). Considering the above-mentioned gaps in cement replacement scenarios, only limited environmental savings can be expected.

2.3 Replacement of cement composites by alkali-activated materials

The consequent step in the way toward more sustainable building materials is based on the introduction of a new type of binder: alkali-activated materials often called geopolymers (Farooq et al., 2021; Shehata et al., 2021). However, despite the long-term effort focused on understanding the mechanism of alkali activation and its potential to replace conventional hydraulic binders, several aspects are still not fully elucidated and subjected to extensive ongoing research. In order to contribute to this field, several papers dealing with the potential reuse of industrial waste products should be mentioned.

For the necessary understanding of the hardening mechanism of alkali-activated binders, the following fundamentals need to be described. AAMs are synthesized during the reaction between the alumino-silicate oxides and alkali activator solutions which are responsible for the formation of the partially or fully amorphous polymeric structures with tetrahedral SiO_4 and AlO_4 (Ruiz-Santaquiteria et al., 2013; Sun et al., 2013; van Deventer et al., 2012). The decomposition of Si-O-Al and Al-O-Si bonds is induced by the increase in pH due to the application of alkaline solutions. Such decomposed groups are consequently transformed to a colloid phase and afterward accumulated and formed into a coagulated structure which results in the formation of a dense and hardened structure of the final material. The created hydration products can be assigned to the characteristics of used precursors; however, the type and molarity of applied alkali activators were together with a variety of curing conditions reported as important variables for a detailed understanding of the alkali-activation process (Pacheco-Torgal et al., 2008). Table 4 describes widely studied constituents of alkali-activated materials together with the achieved compressive strength.

Table 4. Compressive strength of investigated alkali-activated materials

Precursor	**Activator**	**Compressive strength (MPa)**	**Reference**
Fly ash	SS + SH	40.42	Hadi et al. (2018)
Fly ash	SS + SH	52.75	Bhutta et al. (2019)
Fly ash + blast furnace slag	SH	74.40	Alrefaei et al. (2019)
Fly ash + blast furnace slag + silica fume	SS + SH	47.12	Elyamany et al. (2018)
Fly ash + cement	SS + SH	76.51	Zailani et al. (2020)
Clay	SS + SH	47.74	Bayiha et al. (2019)

Metakaolin + blast furnace slag	SS + SH	47.84	Huseien et al. (2018)
Metakaolin + rice husk ash	SS + SH	56.63	Liang et al. (2019)
Blast furnace slag	SH	36.23	Zannerni et al. (2020)
Blast furnace slag	SS + SH	64.62	Bondar et al. (2018)
Blast furnace slag + rice husk ash	SS + SH	59.70	Mehta and Siddique (2018)
Metakaolin + fly ash + silica fume	PS + PH	80.72	Zhang et al. (2020)
Brick dust	SS + SH	46.57	Robayo-Salazar et al. (2017)
Red mud	SS + SH	39.54	Singh et al. (2020)
Rice husk ash	SS + SH	49.65	Nazari et al. (2011)

*SS – sodium silicate; SH – sodium hydroxide; PS – potassium silicate; PH – potassium hydroxide

According to the study by Puertas et al. (2008), different hydration products can be found in alkali-activated pastes. The structure of alkali-activated materials is influenced by the content of calcium oxide, while high calcium materials and low calcium materials can be distinguished (Duxson et al., 2005). AAM with high calcium content is formed mostly by C-A-S-H gel (CaO-Al_2O_3-SiO_2-H_2O) similar to tobermorite, while composites with low calcium content such as metakaolin, brick waste of certain types of fly ashes led to the formation of alkali aluminosilicate gel N-A-S-H (Na_2O-Al_2O_3-SiO_2-H_2O) with pseudo-zeolitic structure while the formation of three-dimensional N-A-S-H gel is induced by the use of sodium activator (Bernal et al., 2010; Provis et al., 2005; Provis et al., 2015). From the point of view of nanostructure and mineralogical characterization of the hydrated products, Ruiz-Santaquiteria et al. (2012) revealed the formation of kaolinite with a minority of illite, muscovite and crystalline montmorillonite in the alkali-activated fly ash obtained from the electric power plant. This type of gel contributes to the final mechanical and durability properties (Balczar et al., 2015; Sun and Wang, 2015), although the zeolitic crystals can be distinguished in the second period of hydration of alkaline activated materials (Lee et al., 2016). The low calcium AAM can be denoted as a subset of AAMs usually named geopolymers. In order to more detail, the distinction of low calcium AAMs, Provis et. al. (2015) revealed the influence of Si/Al ration on resulting properties when Si-poor (Si/Al < 1) or Si-rich (Si/Al > 5) materials are not suitable for the structural construction applications because of low mechanical properties and resistance to salt attack.

As mentioned above, the type of original material as well as its chemical composition is fundamental for the formation of final hydration products. Several authors studied different combinations of used materials in detail, and alkali activators and introduced several models of alkali activation including SiO_2/Al_2O_3 ratios, applied alkali activator, chemical composition, curing temperature, curing age, and precursors. For example, Shi et al. (2011) distinguished several hydration products in dependence on MgO content in blast furnace slag and revealed a whole range and combinations of different hydration products linked to the different mechanical performances. Hydration products of alkali-activated fly ash were studied through SEM analysis and nano-indentation by Němeček et al. (2011), who found at least four different phases. However, despite many performed investigations, some contradictions were described by Granizo et al. (2007), who based on the results of Palomo et al. (1999) focused on different reactions concerning the type of used alkaline activators.

Several barriers have been identified including a wide range of precursors with the variable chemical composition of material inputs, not clearly eluded effect of alkaline activators, limited knowledge about long-term durability, etc. On the other hand, achieved results in terms of mechanical strength refer to the potential to replace the cement-based binders in several applications. Moreover, substantial advances in the preservation of the natural environment are associated with the AAMs application.

Besides the type and composition of a used precursor, several authors highlighted the importance of the fineness of used materials. Considering the conclusions of recently performed studies, very fine particles with a high specific surface can promote the alkali-activation process and improve mechanical strength. Brough and Atkinson (2002) reported that the increase in Blaine fineness from 3300 to 5500 cm^2/g could induce an increase in compressive strength from 65 to 100 MPa. On the other hand, this observation was denied by several other authors who neglected the influence of Blaine's fineness of particles and assigned obtained improvements to the molarity of applied activators (Ken et al., 2015; Reig et al., 2014).

However, considering the advancement in the assessment of modern building materials, functional performance by the meaning of mechanical strength, and durability go hand in hand with requirements paid on the lowered environmental burden, preservation of natural resources, and waste production minimalization (Cong and Cheng, 2021). On top of that, such designed materials must provide sufficient economic viability. In this regard, labeling the alkali-activated materials as sustainable building materials was questioned due to the consumption of commercial alkaline activators (sodium hydroxide, sodium silicate, potassium hydroxide) (Konig et al., 2021).

The effect of the used alkali activators on the microstructure formation poses a fundamental knowledge for a detailed understanding of AAMs. The study performed by Komljenovic et al. (2010) revealed the effect of applied alkali activators (type, concentration, and silicate modulus) on the alkali activation process and thus final material properties. These findings were also confirmed by observations of Tuan et al. (2018) and Yuan et al. (2016), who point to the significant influence of the molar concentration of alkali activators on the mechanical properties of such formed materials. With regards to performed works focused on the combination of alkali activators, pure water glass or in combination with sodium hydroxide represents the most promising compressive strength results (Ken et al., 2015).

Among others, the effect of the curing condition was also widely studied by various authors. Elevated temperature is, in general, considered a critical factor for the acceleration of the microstructure formation and improvement of mechanical parameters. On this account, Yuan et al. (2016) concluded that increased temperature curing has an effect on the formation of more dense structures and increased mechanical strength. Similar findings were noted by Gebregziabiher et al. (2016) however, the modified curing condition in terms of elevated temperature may induce crack formation and deterioration at later ages (Komnitsas et al., 2015; Lee et al., 2016; Luna-Galiano et al., 2016).

Taking into account the completely omitted consumption of Portland cement and concurrent valorization of various waste products, substantial environmental savings can be expected. The environmental analysis aimed at the determination of carbon dioxide as well as energy-saving performed by Amran et al. (2020) carbon point to potential savings of about 45 %, and 70 % respectively. Similar outcomes can be recognized in many other studies (Salas et al., 2018; Robayo-Salazar et al., 2017) who refer to CO_2 saving in 20 to 75% span depending on the used precursor On the other hand, the environmental protection of geopolymers was questioned due to extensive consumption of used alkaline activators as well as the transportation distances.

The prevailing sustainable assessment of innovative building materials is usually based on the estimation of directly emitted carbon dioxide within the material manufacturing as the simplified variant. In more advanced models, carbon footprint analysis or embodied energy analysis are performed to access the environmental footprint in selected boundary conditions. Here, mostly cradle-to-gate or cradle-to-grave boundary conditions are considered. In these terms, only carbon dioxide is intended as an environmental burden, while the rest of the externalities are not involved.

3 Selected publications of the author and their contribution to the development and assessment of sustainable cement- and geopolymer composites

The adverse effects on human activities associated with the construction industry pose a major challenge for modern society in terms of the preservation of natural resources and biodiversity. However, despite the enormous effort of researchers, this issue is still fully resolved and suitability principles are not fully adopted at the required scale. This work summarizes the major contribution of the author to the development and assessment of innovative building materials heading to truly sustainable products which integrate functional, environmental, and economic requirements. Primarily motivation which should be intertwined with all listed research papers lies in exploring the potential of waste materials, identification of major barriers, breaking unsatisfactory paradigms, and integrating assessment criteria for building materials.

The following part of the work is divided into 5 subsections, in which the most significant research papers related to the area are provided in full text.

3.1 Replacement of cement by supplementary cementitious materials

Selected journal paper

Fort, J., Sal, J., Sevcik, R., Dolezelova, M., Keppert, M., Jerman, M., Zaleska, M., Stehel, V., Cerny, R., 2021. Biomass fly ash as an alternative to coal fly ash in blended cements: Functional aspects. *Construction and Building Materials 271, 121544.*

In this paper, the commonly used cement replacement – coal combustion fly ash is replaced by wood-based fly ash, consequently, chemical characteristics and functional parameters are described in detail. The primary motivation lies in future abatement (and thus limited availability of CFA) as well as the mitigation of environmental hazards associated with the presence of heavy metals in CFA. Complex characterization of biomass fly ash (BFA) performed prior to the other investigations reveals very good prerequisites for its use as SCM. The experimental analysis of functional properties of Portland cement-BFA-based composites after 28-days curing shows the suitability of BFA as SCM for cement replacement up to 30 % by mass. The significantly lower content of hazardous elements in BFA compared to CFA provides the benefit in form of possible application of such material in blended cement without any further processing or treatment.

Contribution to the practice: This paper contributes to the exploration of the potential of SCMs as a replacement for CFA for the manufacturing of „green“ cement in the sense of reduced CO_2 *emissions as well as lowered heavy metals content.*

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Biomass fly ash as an alternative to coal fly ash in blended cements: Functional aspects

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HIGHLIGHTS

- The concentration of hazardous elements in biomass fly ash is substantially lower compared to coal fly ash.
- As for the functional properties, biomass fly ash can replace up to 30% of Portland cement in composites.
- Biomass fly ash without any preprocessing can serve as a future replacement of coal fly ash in blended cements.

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ABSTRACT

The tightening of environmental standards in the EU including the presupposed abatement of coal combustion power plants presents an important impulse towards the transition to a circular economy. However, the achievement of this challenging goal requires a solution to a number of problems. One of the downstream problems consists in the fact that the application of fly ash originating in coal combustion (CFA) as a partial cement replacement might be gradually abandoned in future decades. The first reason may be economical as the increasing environmental regulations will probably result in its more costly processing. The second one is related to the increasing use of alternative energy sources which can lead to the decreasing availability of CFA. Biomass fly ash (BFA) originating from wood combustion can be considered as one of the prospective environmentally more friendly candidates for a partial replacement of CFA as supplementary cementitious material (SCM). In this paper, functional aspects of a possible replacement of CFA by BFA in blended cements are analyzed. Complex characterization of BFA performed prior to the other investigations reveals very good prerequisites for its use as SCM. The experimental analysis of functional properties of Portland cement-BFA based composites after 28-days curing shows the suitability of BFA as SCM for the cement replacement up to 30% by mass. The data collected after 90 and 180 days of curing indicate a substantial improvement of strength of all analyzed composites. The significantly lower content of hazardous elements in BFA in a comparison with CFA presents another benefit; it can be used in blended cements without any further processing or treatment.

1. Introduction

Environmental aspects of human activities pose a big challenge for the present society. As a consequence of commitments towards decarbonization of industry and mitigation of greenhouse gasses production, the concrete industry faces criticism related to excessive production of carbon dioxide [7,70]. Moreover, cement production is associated with depletion of natural resources which do not comply with the principles of the circular economy [2]. The replacement of traditional Portland clinker has been found as one of the opportune strategies for the reduction of negative environmental externalities associated with cement manufacturing [4,11]. On this account, several types of pozzolanic materials also known as supplementary cementitious materials (SCMs) have been extensively studied over the past decades [17,20,29,36,37,46, 49, 50,64,71–73].

Fly ash (FA) originating in combustion power plants has been recognized as a very efficient and abundant material for partial cement replacement [3,37,44,55,67]. However, its use in blended

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cements can be problematic soon due to the tightening of environmental standards in the EU. Coal fly ash (CFA) and municipal solid waste incineration ash (MSWIA) are major anthropogenic sources of heavy metals, such as Hg, B, Cd, As, Se, Pb, Cr, Ni, Mn, Co, Sb, V, Cu, and Zn. In addition, CFA may contain organic pollutants and radionuclides [32].

Pretreatment techniques for mitigation of potential environmental hazards related to the utilization of CFA involve bioleaching, chemical leaching, physical leaching, thermal treatment, electrochemical methods and their combination [32,45]. However, these processes can affect the eventual environmental benefits associated with blended cements [74]. Additionally, in the light of increased awareness and commitments to climate change, CO_2 emission, in particular, coal combustion became an undesired source of primary energy [79]. Since coal combustion power plants generate a substantial share of CO_2 production (also SO_2 and NO_x), their operation may be gradually reduced in the near future. The decrease in CO_2 emissions goes hand in hand with the abatement of coal combustion power plants which was already demonstrated in several countries. In the United States, the amount of coal used for energy production dropped by 50% in the last 10 years [2]. In the European Union, GHG emissions should be cut to almost zero by 2050 [7,26,61]. The mid-term and long-term energy strategies rely on moving away from traditional coal combustion towards renewable energy sources [41,63]. As a consequence, CFA may become scarce compared to the present and its replacement by other available and suitable SCMs may turn into a necessity [12].

Biomass power plants contribute with 16% to the gross final energy consumption for heating and cooling, electricity production and transport in the EU and a further increase up to 33 – 50% is expected to meet the renewable energy action plans [48]. The waste products of biomass combustion can be considered as one of the prospective SCMs which can be utilized in the construction industry and replace some of the currently used pozzolans [43,51] while bringing environmental benefits [19,27]. Their application in blended cements was studied in countries with dominant agroindustry in particular, where such utilization of agricultural waste products (e.g., rice husk, bagasse or palm oil fuel ash) has great potential because of their abundance. Nonetheless, the obtained findings are valuable only for the countries and regions where the cultivation of these crops is possible. Despite the increasing number of studies dealing with the valorization of biomass fly ash (BFA), which appeared during the last decade or two, the obtained knowledge could not be generalized yet. The results of such analyses were always related to the source and type of applied BFA [29,62]. In the EU, any application of BFA as SCM has to take into account that particular countries have a tradition in the cultivation of very different crops with different chemical compositions [40,72]. Taking into account the abundance of BFA sources, wood waste is considered as a prospective alternative since 70% of WBA remains landfilled. Obviously, some recommendations can be given, e.g. that finer BFA particles are more suitable for cement replacement than coarser ones from the point of view of the resulting strength [35,50]. As noted by Medina et al. [44], a partial replacement of cement by biomass fly ash (up to 20 wt %) still provides a satisfactory level of functional parameters. However, more pronounced environmental benefits are associated with larger volume applications [69]. The main drawback of this material consists in the presence of numerous organic and inorganic compounds causing a significant diversity in both chemical and phase composition. This variety is related not only to the type of the combusted material but also to the applied technology. General guidelines are thus very difficult to formulate [5]. Notwithstanding, trial studies performed, e.g., by Carevic et al. [10], del Bosque et al. [23], and Rissanen et al. [58] provided an overview of WBA potential in the field of cement replacement. While the most of studied WBA is being viewed as suitable considering the pozzolanic and hydraulic activity thanks to CaO-Al_2O_3-SiO_2 diagram, a major weakness consists in the relatively high content of undesired compounds, such as alkalis, CaO, MgO that refer to possible risks of material degradation as a consequence of material swelling and cracking [9,31]. The effect of free CaO and MgO was discussed also in a relation to the increased water requirements that affected the morphology of designed mixtures [54]. Compared to coal fly ash based composites, different crystalline minerals were found, including calcium chlorides and phosphates.

The main aim of this study is to analyze functional aspects of the application of BFA as an alternative to CFA in blended cements. The research includes experimental analysis of chemical and physical properties of BFA and its consequent utilization as eco-friendly SCM in mortars. The effect of applied BFA on functional properties at cement replacement dosages ranging from 10% to 70% by mass is described using isothermal calorimetry, X-ray fluorescence (XRF), X-ray diffraction (XRD), scanning electron microscopy (SEM), mercury intrusion porosimetry (MIP), helium pycnometry, and mechanical tests.

2. Materials

2.1. Biomass fly ash

The wood-based biomass fly ash was collected from the biomass electricity plant in Trutnov, Czech Republic. This facility uses a fluidized bed furnace for the combustion of wood chips composed mainly of softwoods, with only a minor share of hardwoods. The annual BFA production of this facility reaches 80 kilotons and most of the generated ash serves as landscaping element due to the lacking available utilization options.

The combustion process of wood chips/pellets is composed of several consequent reactions such as drying, devolatilization, gasification, combustion and gas-phase reactions. An informative description is given in Fig. 1. Heating of the wood chips used caused hydrolyzation, dehydration, and oxidization that create the formation of combustible volatile substances along with the temperature increase. Reaching the ignition temperature of the volatiles and tarry substances begin the combustion process that results in the decomposition of cellulose, hemicellulose, and lignin. The fluidization flow regime with velocity from 4 to 10 m/s was maintained (Fig. 2).

The chemical composition of BFA was examined by the X-ray fluorescence (XRF) spectroscopy (Thermo ARL 9400 XP). The phase composition was studied by the X-ray diffraction (XRD) method (PANalytical X́PertPRO diffractometer equipped by CoKα X-ray tube, 40 kV, 30 mA). The Rietveld refinement of experimental data was performed by the help of the BGMN code. The amorphous portion was quantified by the help of internal standard; fully crystalline zincite (ZnO) was added to samples (20% by mass) and the obtained pattern was refined again. The amount of amorphous matter was calculated on the basis of the known amount of ZnO in the matter by BGMN code.

The XRF analysis showed SiO_2, CaO, and Al_2O_3 as major oxide compounds; together they comprised 88% wt.% of the BFA powder. This finding was in accordance with Maschowski et al. [42] and Medina et al. [44] who came to the conclusion that a dominant content of CaO and SiO_2 can be expected from wood biomass combustion. Moreover, the sum of SiO_2, CaO, and Al_2O_3 exceeding 70 wt% is considered as a threshold value for the material suitability as a pozzolan admixture. A minor content of Fe_2O_3, K_2O, MgO, P_2O_5, MnO, and Na_2O was traced. The increased content of water-soluble compounds as chlorides and sulfates can be viewed as one of the weaknesses of the applied BFA since an increase of

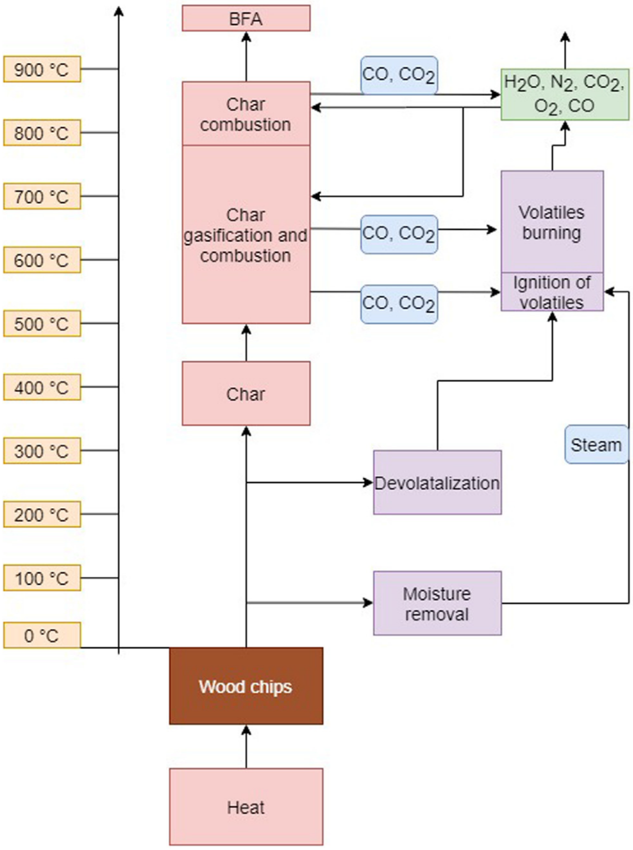

Fig. 1. Simplified scheme of biomass combustion process.

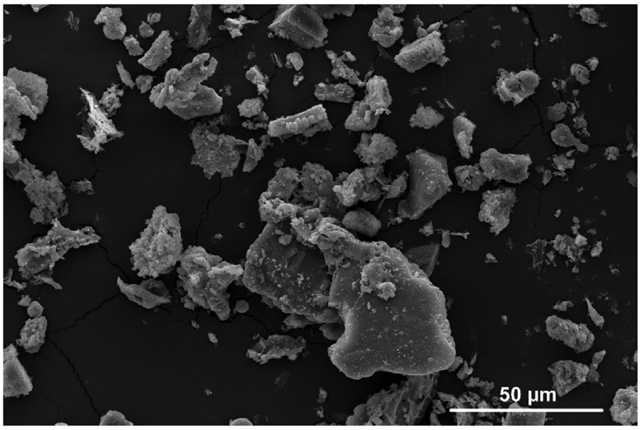

Fig. 2. SEM image of raw BFA.

porosity and consequent decrease in strength may occur. In particular, the possible negative effect of precipitation of ettringite or brucite represents a major concern due to their expansive character [10]. The detailed chemical composition of the studied BFA is given in Table 1.

The mineralogical composition of BFA is presented in Table 2 via XRD and consequent Rietveld refinement. The amorphous phase content was dominant, having an almost 53 wt% share. Among the crystalline phases, quartz (SiO_2) and portlandite ($Ca(OH)_2$) were identified as the most represented. Nevertheless, the quartz content was relatively low, considering the chemical composition of BFA listed in Table 1 and also the results obtained by Rissanen et al. [57]. The apparent reason was a high content of SiO_2 in the amorphous matter that can be viewed as a positive prerequisite for the hardened structure formation and thus satisfactory mechanical performance. Calcite ($CaCO_3$), lime (CaO), muscovite ($KAl_2(AlSi_3O_{10})(OH)_2$), and anorthite ($CaAl_2Si_2O_8$) were found in minor amounts. The revealed muscovite content can be viewed as an undesired impurity that is probably accompanied by the contamination of processed waste wood by other agricultural residues such as rape straw that may contain a small amount of clay. Alternatively, the wood chips collection and processing by using heavy machinery may result in possible contamination of ash by clay minerals. Fortunately, muscovite is viewed as a high-temperature resistant mineral that does not exhibit any distinct changes below 1 000 °C since the dehydroxylation that may occur in the range of 900 – 1000 °C depends on the structure and crystal size [28]. In this sense, muscovite serves as filler next to used aggregates.

Thanks to the notable calcium content in the analyzed BFA, at least partial hydraulic properties could be expected [40].

The composition of BFA can vary significantly, and its strongly dependent on the type of the used biomass source [68]. Nonetheless, substantially lower concentrations of hazardous compounds, such as heavy metals, may be expected in a comparison with CFA, which begins to be considered as an environmental hazard worldwide. Jambhulkar et al. [32] reported that CFA disposal struggles with bioaccumulation, as hazardous elements can enter the food chain and have toxic effects for humans.

In order to assess the potential risks, the comparison of the studied BFA with the averaged values measured for CFA originated in EU28 [52] and the data obtained from three coal combustion power plants in Poland [8] is given in Table 3. The limiting values for hazardous elements in building materials are not available but for a basic assessment of the health and environmental risks associated with fly ash utilization the limits for soils can be used instead. The cadmium content of 0.5 mg/kg is viewed as a threshold whose exceeding can affect cell proliferation and differentiation, thus cause mutations and chromosomal deletions potentially [13,38]. The exceeded limit for chronic arsenic exposure of about 50 mg/kg increases the risk of cancer, heart diseases, and night blindness [66]. Substantial negative effects can be related also to the chromium content due to its poisonous effect especially in the case of hexavalent chromium that is hemotoxic, genotoxic, and carcinogenic [75]. Unfortunately, the potential risk and negative health consequences including the threshold limits is not described in detail for particular elements. Notwithstanding, the precautionary principle should be taken into account to avoid major health issues [77]. Apparently, the concentration of all potentially harmful elements in the BFA used in this paper was significantly lower compared to CFA, in some cases it was not even detectable. Therefore, contrary to CFA, the utilization of the analyzed BFA did not require any intensive treatment for immobiliza-

Table 1
Chemical composition of raw BFA.

Compound	SiO_2	CaO	Al_2O_3	SO_3	Fe_2O_3	K_2O	MgO	P_2O_5	MnO	Na_2O	Cl
wt.%	45.3	19.9	13.0	4.7	4.7	4.3	2.7	1.7	2.7	0.8	0.2

Table 2
Phase composition of raw BFA.

Phase	wt.%
Amorphous	52.9
Quartz (SiO_2)	11.2
Calcite ($CaCO_3$)	7.6
Lime (CaO)	6.1
Portlandite ($Ca(OH)_2$)	11.9
Anorthite ($CaAl_2Si_2O_8$)	2.7
Muscovite (KAl_2 ($AlSi_3O_{10}$) $(OH)_2$)	3.9
Anhydride ($CaSO_4$)	3.7

Table 3
Comparison of hazardous elements in CFA and BFA (in mg/kg).

	Moreno et al. (38)	Bradlo et al. (39)	BFA (this study)		
	EU fly ashes	P1	P2	P3	
As	51	22	39.8	68.8	n.d.
B	259	125	144.3	179.9	n.d.
Ba	1302	–	–	–	139
Be	8	–	–	–	n.d.
Cd	2	–	–	–	n.d.
Co	35	–	–	–	n.d.
Cr	148	81.8	100.4	91	8.2
Cu	86	51.8	86.1	167.1	10
Ge	7	–	–	–	n.d.
Hg	0.2	–	–	–	n.d.
Li	185	–	–	–	n.d.
Mo	11	5.5	8.5	10.9	n.d.
Ni	96	55.7	81.9	113.4	n.d.
Pb	80	–	–	–	16.4
Rb	108	–	–	–	7.7
Sb	4	–	–	–	n.d.
Se	7	–	–	–	n.d.
Sn	8	–	–	–	n.d.
Sr	757	–	–	–	38
Th	30	–	–	–	n.d.
U	12	–	–	–	n.d.
V	228	–	–	–	106
Zn	154	124.8	229.2	483.3	11.4
Ga	–	55.2	76.9	117.9	n.d.
La	–	14.8	18.75	32.28	n.d.
Ce	–	30.2	33.1	49.4	n.d.
Pr	–	3.5	4	6.5	n.d.
Nd	–	15.17	17.68	31.12	n.d.
Y	–	17	20.34	23.7	n.d.

n.d. – not detected.

tion of hazardous substances and provided a more favorable alternative, as for the environmental concerns [76].

The microstructure of BFA samples was observed using scanning electron microscopy (SEM), using an FEI QUANTA FEG 450 instrument equipped with a backscatter electron, secondary electron, and energy dispersive (EDS) detectors. The samples were coated with 5 nm thick gold film prior to the analysis and the experiments were done at 20 kV accelerating voltage.

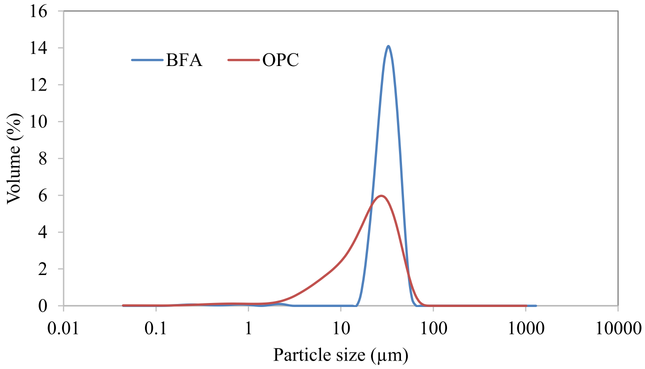

Fig. 3. Particle size distribution of OPC and raw BFA.

The micrograph of raw BFA is given in Fig. 1. BFA consisted mainly of a heterogeneous mixture of particles with high variety of morphologies and sizes. The size of particles ranged from tens of nanometers op to tens of micrometer. As depicted in Fig. 1, most of the particles is present as larger microsized aggregates with rugged surfaces. Compared to the microstructure of CFA, the irregularity of BFA particles could increase the interparticle friction forces during the mixing period and thus negatively affect the workability of the fresh mixture [6].

The pozzolanic activity of raw BFA was determined by the Chapelle test according to the NF P18-513 standard. The measurement was based on the quantification of $Ca(OH)_2$ fixed by 1 g of BFA when mixed with 2 g of CaO and 250 ml of distilled water [78]. The mixture was heated up to 80 °C for 24 h with continuous stirring. Consequently, the samples were filtered with the Bünner funnel. The filtrate was titrated against HCl with bromophenol blue indicator to determine OH^- concentration. The Ca^{2+} concentration was obtained by titration with EDTA solution using the Murexide indicator. The results of the Chapelle test showed for the analyzed BFA 606 mg CaO/g, substantially exceeding the minimum value of 330 mg CaO/g. Similar values were obtained, e.g., for calcareous fly ash or siliceous fly ash from coal combustion [39]. The measured CaO concentration was even higher compared to sugarcane bagasse ash [18] or bamboo stem ash [59]. Therefore, the studied raw BFA could be considered as a good pozzolan, which made good prerequisites for its effective utilization in cement-based materials.

The particle size distribution of BFA was determined using an Analysette 22 MicroTec plus (Fritsch) laser diffraction device having a measuring range of 0.02 μm to 2 mm. Apparently, the analyzed BFA did not require any further treatment, as for the particle fineness (d_{50} = 32.7 μm) which was similar to OPC (Fig. 3), and could be easily incorporated into cementitious materials.

2.2. Composite mixtures

Portland cement CEM I 42.5 R meeting the requirements of the ČSN EN 197–1 standard was used as the main binder. The dosage of

Table 4
Composition of studied mixtures.

Mixture	Cement (g)	BFA (g)	Aggregate (g)	w/b	Soundness (mm)	Setting time initial/final (min)
REF	450	0	1350	0.50	3	160/230
BFA10	405	45	1350	0.50	2.5	185/255
BFA20	360	90	1350	0.50	4	190/250
BFA30	315	135	1350	0.56	3.5	210/275
BFA40	270	180	1350	0.56	4	240/315
BFA50	225	225	1350	0.61	7.5	250/330
BFA60	180	270	1350	0.61	8.5	285/375
BFA70	135	315	1350	0.67	10	305/420

BFA as partial cement replacement was up to 70 wt% in 10 wt% steps. Standard silica sand with continuous granulometry (PG1-PG2-PG3) was used as aggregate. The amount of water in the mixes expressed by the water/binder (w/b) ratio corresponded to the spread diameter of 150 mm. Detailed information on the composition of the mixtures is given in Table 4. The soundness was determined according to EN 196–3. Since the content of free CaO and MgO may present a risk of swelling, all tested mixtures were tested by the Le Chateliers rings according to EN 450–1 standard with satisfactory results meeting the criteria of maximum distance between the needle ends. The significant differences in initial and final setting times refer to an adverse effect of alkalis in BFA [10].

3. Experimental methods

The phase composition and microstructure of hardened composite mixtures was analyzed by XRD and SEM using the methodology and devices described in Section 2.1.

The effect of BFA on material porosity and pore size distribution was studied by Mercury Intrusion Porosimetry (MIP) with the help of Pascal 140 and Pascal 440 porosimeters (Thermo Scientific). At the evaluation of measured data, the circular cross-section of capillaries was assumed, whereas the mercury contact angle was 130°.

Bulk density, matrix density and total open porosity of designed mixtures in the hardened state were determined as representatives of basic physical properties. The bulk density was obtained by weighing of dried samples and determination of their volume using a digital caliper. For the measurement of matrix density a helium pycnometry device Pycnomatic ATC (Thermo Scientific) was used. The total open porosity was calculated on the basis of the known bulk and matrix densities.

The isothermal heat flow calorimeter KC 01 was applied for the measurement of hydration heat development in the studied mixtures [33]. The measurement was carried out with a 1 g sample of a dry mixture of each mixture in a cylindrical copper vessel having a diameter of 8.25 mm and height 70 mm. The calorimeter with the solid sample and liquid components operated at a temperature of 25 °C. After about 1 h of stabilization, the calorimeter was opened and water was injected into the vessel. Consequently, the paste was mixed for 30 s by rotation of the plastic tubule mixer, and the vessel was locked by a rubber plug.

The compressive and flexural strength was determined using a hydraulic testing device Servoplus Evolution (Matest) having a stiff loading frame with the capacity up to 6000 kN. The experiments were carried out after 28, 90, and 180 days of curing. Before the experiments, the samples were placed into an oven and dried at 80 °C for 48 h.

The strength activity index (SAI) was calculated to assess the functional performance of the applied BFA. SAI is a widely used indicator for the assessment of SCM applications. Compared to the Chapelle test, the SAI index is based on a comparison of the compressive strength of particular mixtures to the reference Portland cement paste. However, this approach is often criticized for its inaccuracy [30]. Moreover, this indicator provides only a qualitative evaluation without any details. Therefore, for a more precise assessment the pozzolan effectiveness coefficient (PEC) introduced in Zaleska et al. [78] was used. PEC is defined as follows:

$$\mathrm{PEC} = \frac{f_c(Px) - \left(1 - \frac{x}{100}\right) f_c(R)}{f_c(R) \cdot x/100}$$

where f_c (MPa) is the compressive strength, x the content of pozzolanic admixture in wt.%, Px the paste prepared by the mixture of cement and x% of the pozzolan as a cement replacement, R the Portland cement reference paste. The evaluation of PEC results is based on the following classification: i) 0 < PEC < 1 – the applied admixture can be considered as pozzolan active, higher PEC value means higher pozzolanic activity.; ii) PEC < 0 – the applied material can be considered only as a filler without any pozzolanic activity; iii) PEC > 1 – the effectiveness of the pozzolan is higher than of Portland cement, which indicates synergetic effects in the Portland cement-pozzolan-water system.

4. Results and discussion

4.1. Phase composition and microstructure

An example of measured XRD patterns is given in Fig. 4 (BFA 30 mortar) where quartz, calcite, anorthite, and muscovite were identified as the main crystalline components of biomass containing mortars. The results of the quantitative XRD analysis of all analyzed composites are presented in Fig. 5. The decreasing amount of portlandite with the increasing BFA content which was observed up to 40% of BFA in the blended binder was related to the extent of the pozzolanic reactions in the particular mixes. The increase of the content of quartz, muscovite, calcite, and feldspar with the increas-

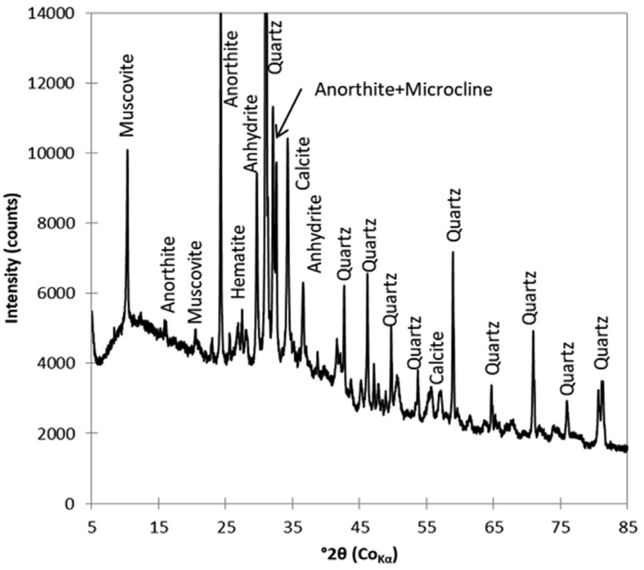

Fig. 4. XRD pattern of BFA 30.

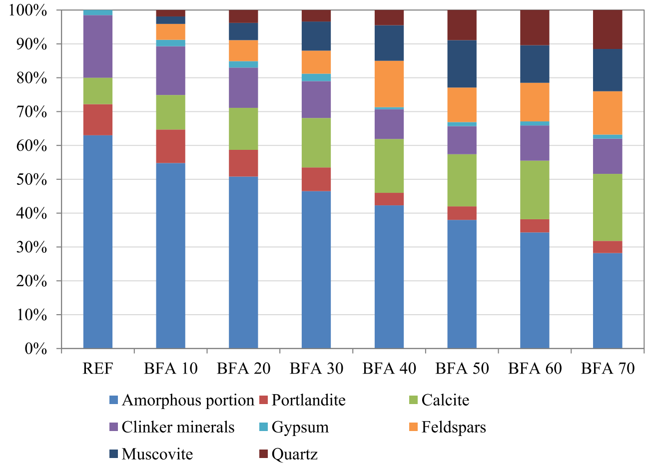

Fig. 5. Phase composition of studied mortars after 28 days.

ing BFA dosage indicated a presence of nonreacted BFA which became apparent for the mixes with the cement replacement level of 40% and higher, in particular. The decreasing share of the amorphous phase in the hardened mixes reflected then the decreasing amount of CSH gels.

SEM micrographs of hardened composites after 28 days of curing are presented in Fig. 5. Apparently, the microstructure of the composites started to be more complex with the increased quantity of incorporated BFA. In addition, the increased amounts of BFA resulted in a network containing more pores. Based on the morphology of particles reported elsewhere [4,14,16], some of the main phases were identified. In Fig. 6A (reference sample), spherical particles of clinker mineral, belite, were identified, together with the scalenohedral calcite and CSH. Scalenohedral calcite was formed probably due to the excess of Ca^{2+} cations, as reported by Cizer et al. [16]. In contrast to the reference sample, crystals of gypsum and typical rhombohedral crystals of calcite were identified in BFA 10 (Fig. 6B). In BFA 20 (Fig. 6C), needle-like crystals of ettringite, a product of cement hydration with chemical formula $Ca_6Al_2(SO_4)_3(OH)_{12}{\cdot}26H_2O$, were detected. This phase, which may promote the deterioration of the mortars and was already observed by other authors [56], was though presented only in negligible quantities. This observation was in an agreement with XRD result where no diffraction peaks of ettringite were found in the XRD patterns (see Fig. 4 and summarization of quantitative phase analysis in Fig. 5). As it was discussed above, due to

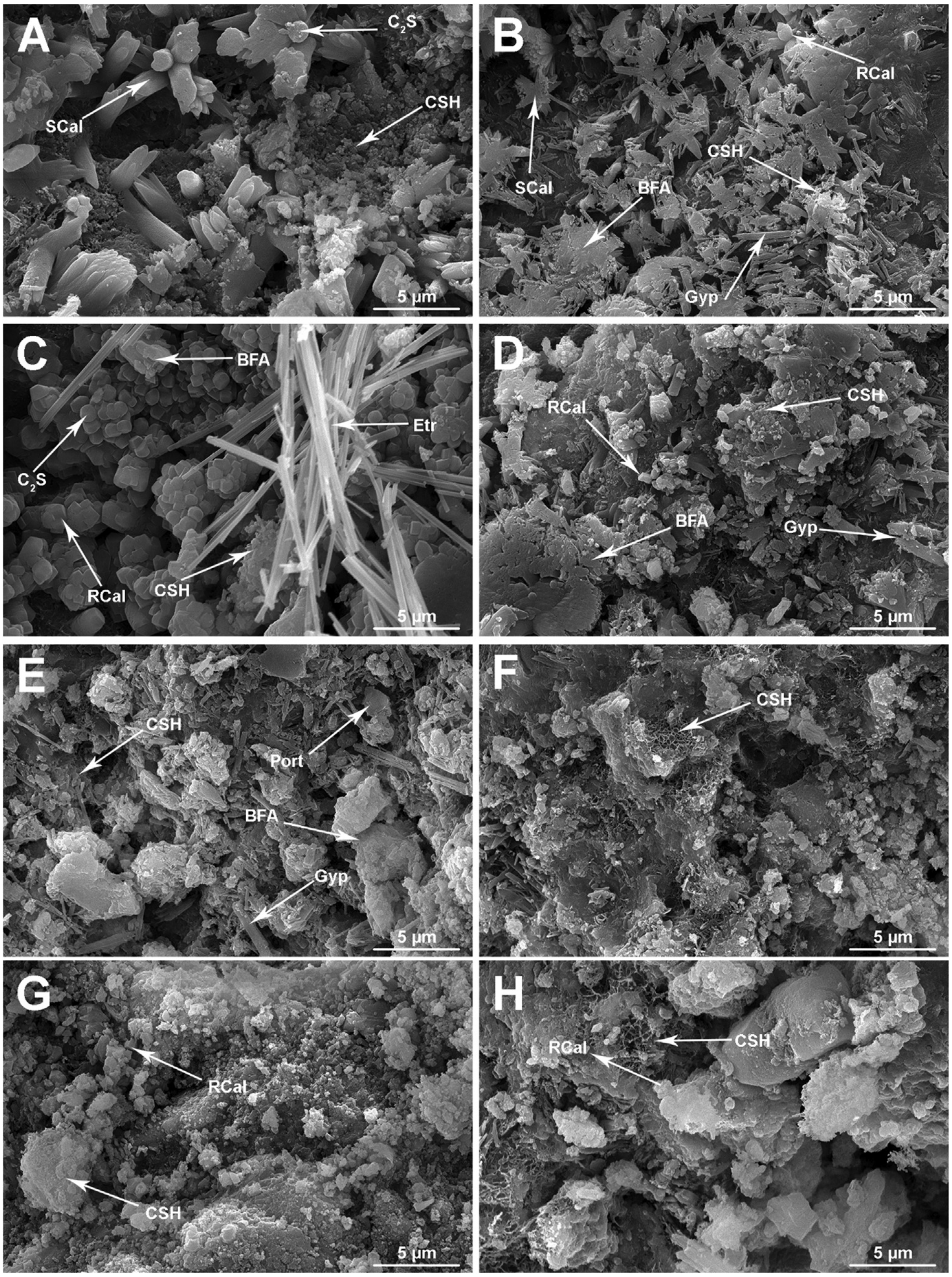

Fig. 6. SEM micrographs of studied composites after 28 days of curing (A - REF, B - BFA 10, C – BFA 20, D – BFA 30, E – BFA 40, F – BFA 50, G – BFA 60, H – BFA 70) with the identification of some of the main phases (SCal – scalenohedral calcite, C_2S – belite, CSH – calcium silicate hydrate, RCal – rhombohedral calcite, BFA – biomass fly ash, Gyp – gypsum, Etr – ettringite, Port – portlandite).

the irregularities and high variability of BFA particle morphology, the identification of phases was more demanding in the samples with higher BFA dosage. Some of BFA particles were identified in Fig. 6B–E, together with the partially carbonated portlandite crystals, rhombohedral calcite, gypsum, and CSH. In the BFA 50–70 samples (Fig. 6F–H), BFA particles seemed to be mostly covered by the hydration products, confirming their good chemical compatibility with cement.

4.2. Hydration heat

The specific hydration heat power generated by the studied blended binders is shown in Fig. 7. The first peak at ~ 3 min. after mixing all compounds together can be assigned to C_3A hydration. Additionally, wetting heat also contributed to the first maximum. The second maximum corresponding to the C_3S hydration was observed after ~ 10 h. It gradually decreased in line with the increased amount of applied BFA admixture. While for the reference OPC paste the maximum was about 2.5 mW/g, for the mixture containing 70 wt% of BFA it dropped to 0.7 mW/g. The obtained values refer to the reduction of hydration reaction as a direct result of OPC replacement by BFA. This observation was in accordance with Deboucha et al. [22] who found that the use of 40 wt% of blast furnace slag resulted in a decrease in the hydration heat peak of about 35%. They concluded that latent hydraulic reaction released less heat during hydration compared to OPC. Further explanation can be found in more available water for the hydration of cement when BFA content was increased [47]. The negative effect of utilization of CFA on hydration heat evolution was reported by Snelson et al. [65] who attributed this reduction to the presence of alkalis and chlorides. The decrease in hydration heat evolution can be considered as a potential advantage since lower temperature gradients lead to restriction of thermal contraction and crack formation.

4.3. Basic physical characteristics

The basic physical properties of studied mortars after 28 days of curing are given in Table 5. The observed decrease in bulk density (up to 8% in a comparison with the control specimen) could be attributed to higher water dosages, as a result of reduced workability of the mixtures with BFA. As the matrix density remained basically unaffected by the use of BFA in the mixes, the open porosity increased significantly with the increasing BFA content, from 21.7% for REF to 27.4% for BFA70. The adverse effect of SCMs on materials porosity was reported also for other pozzolans [39,40,60,62].

Fig. 8 shows the development of total open porosity during the time period of 28 to 180 days. A comparison of data obtained for 28 days and 180 days revealed that the porosity decrease with time was more pronounced for mortars with higher BFA dosage, which indicated improved strength parameters at later ages. This finding can be attributed to a slower hydration process within the first 28 days when the small pores in specimens containing pozzolans were not fully filled yet by hydration products [57]. An irregularity was observed for BFA10 which showed a noticeable drop in the material porosity similar to BFA40.

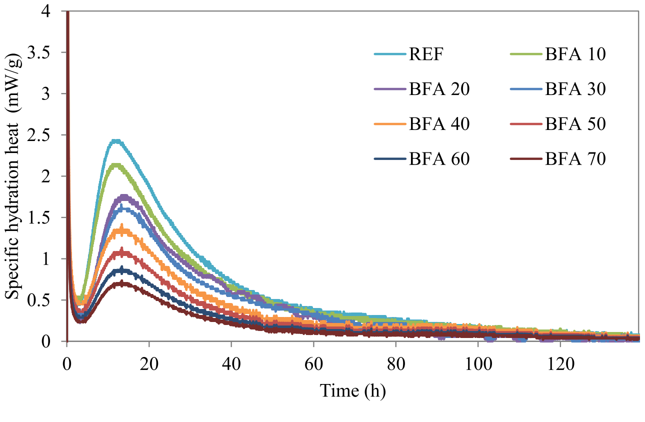

Fig. 7. Evolution of hydration heat in studied blended binders.

It should be noted that the total open porosity of composites based on Portland cement-CFA blends was also higher than of Portland cement mortars but not any changes even after a longer curing period were observed [15].

The results of the MIP experiments (Fig. 9) showed that the incorporation of BFA into the studied mortars affected the pore distribution in a significant way, while a qualitative agreement with the observed changes in total open porosity (Table 5) was

Table 5
Basic physical properties of studied mortars.

Mixture	Bulk density (kg/m^3)	Matrix density (kg/m^3)	Total open porosity (%)
REF	2084 ± 42	2663 ± 17	21.7 ± 1.3
BFA10	2051 ± 40	2691 ± 20	23.8 ± 1.0
BFA20	2040 ± 38	2691 ± 15	24.2 ± 1.7
BFA30	2008 ± 39	2690 ± 16	25.4 ± 2.2
BFA40	1993 ± 32	2675 ± 12	25.5 ± 1.4
BFA50	1978 ± 35	2658 ± 20	25.6 ± 1.9
BFA60	1954 ± 38	2635 ± 19	25.9 ± 1.8
BFA70	1916 ± 33	2637 ± 20	27.4 ± 2.0

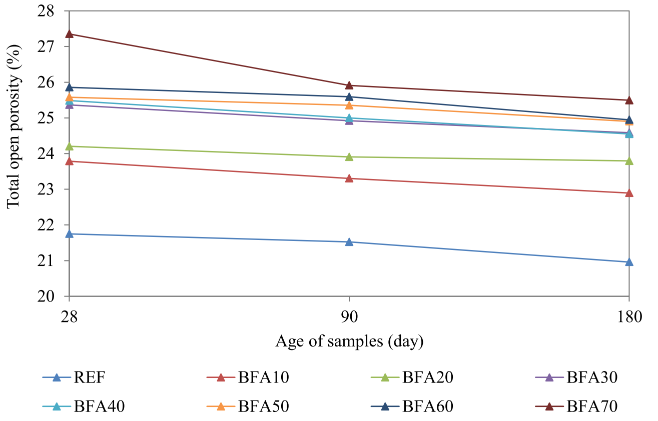

Fig. 8. The effect of sample maturing on total open porosity.

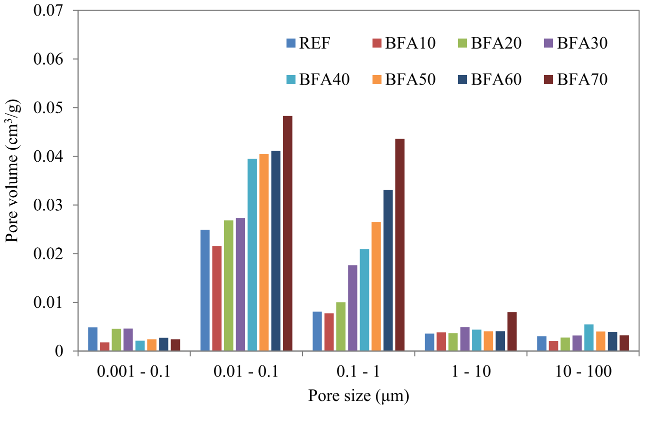

Fig. 9. Pore size distribution of studied mortars.

achieved. The most remarkable differences were observed in the 0.01 μm – 0.1 μm and 0.1 μm – 1 μm ranges where the pore volume increased with the increasing BFA dosage up to two and four times, respectively, in a comparison with the REF samples. This finding was in accordance with Teixeira et al. [69] who concluded that a highly irregular shape of pozzolan particles may increase the number of small pores. The most important information obtained from the MIP measurements was the identification of a threshold in BFA dosage which was 30% by mass.

4.4. Mechanical parameters and pozzolanic activity indices

The compressive and flexural strength of BFA containing mortars after 28 days of curing are given in Table 6. As one can see, a considerable reduction in both compressive and flexural strength was found for mixtures with high BFA content. On the other hand, only a slight decrease in compressive strength was observed for 10 wt% and 20 wt% cement replacement by BFA. A satisfactory level of mechanical performance was maintained up to the BFA content of 30 wt% in the blended cement when the compressive strength was 49.1 MPa and flexural strength 8.1 MPa. The BFA40, BFA50, BFA60, and BFA70 mixes exhibited a more distinct reduction in all mechanical parameters which was in a good agreement with the analysis of phase composition and microstructure of studied mortars (Section 4.1) and the changes in their total open porosity and pore size distribution (Section 4.3). Based on the measurement of basic physical characteristics and mechanical parameters after 28 days of curing, the 30 wt% cement replacement by BFA could thus be considered as a threshold parameter for viable mix design. It should be noted that, in general, the total open porosity correlates closely with the compressive strength despite some rare exceptions which are usually based on a formation of weak binder matrix of alternative SCMs [60]. Notwithstanding, the results obtained in this paper provide more detailed information compared to, e.g., Farinha et al. [25] or Rissanen et al. [57].

Table 6
Mechanical parameters of studied mortars after 28 days.

Mixture	Compressive strength (MPa)	Flexural strength (MPa)
REF	61.7 ± 2.3	9.5 ± 0.6
BFA10	58.2 ± 2.4	8.8 ± 0.7
BFA20	54.3 ± 3.2	8.7 ± 0.8
BFA30	49.1 ± 2.7	8.1 ± 0.6
BFA40	42.7 ± 3.4	8.1 ± 0.3
BFA50	24.4 ± 1.8	6.4 ± 0.5
BFA60	19.4 ± 2.3	4.9 ± 0.3
BFA70	13.1 ± 1.3	4.2 ± 0.4

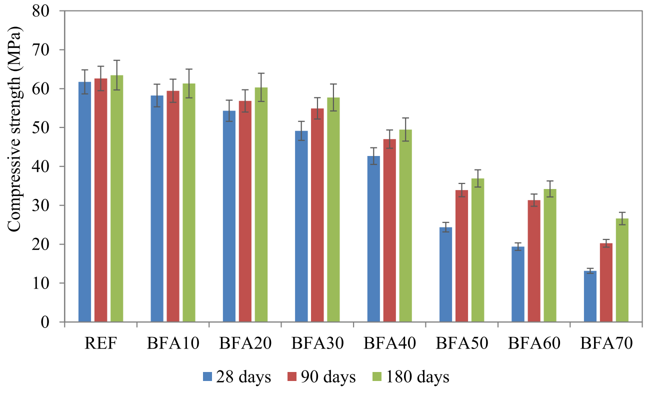

Fig. 10. Development of compressive strength after 28, 90 and 180 days of curing.

The measurements of compressive and flexural strength after 90 and 180 days (Figs. 10, 11) confirmed that the pozzolanic activity can positively affect mechanical parameters at later ages due to the slower course of pozzolanic reaction compared to cement hydration. The densification of the matrix of the analyzed mortars over time resulted in the growth of all strength parameters as is documented in Fig. 12. This improvement was more distinct for mixtures having a higher content of BFA. Specifically, while the compressive strength of the reference mixture after 180 days was improved only by 3%, as compared with 28 days of curing, for BFA70 it was doubled to the final 26.6 MPa. On the other hand, the flexural strength did not exhibit such strength gain and revealed an ~ 20% increase for all mixtures with only slight variations.

The results obtained at later ages did not refer to any adverse effect of the content of water-soluble compounds that may cause undesirable swelling and damage of the hardened structure. These concerns are mainly related to the increased content of alkalis and sulfates as reported, e.g., by Carevic et al. [10], who found a correlation between the increased content of alkalis and decreased compressive strength. In our study, the more pronounced increase in strength parameters, which was observed for mixtures with higher BFA content at later ages, indicated that the presence of reactive compounds could partially compensate the potential negative efects of alkalis and sulfates. On the other hand, a clear beneficial effect of free lime content on the strength parameters which was reported, e.g., by Nawaz et al. [53] was not confirmed in this paper. Deterioration of microstructure observed by some researchers (e.g., Elinwa and Mahmood, 2002) at later ages as a consequence of the expansion due to the alkali-aggregate reaction, was not found either.

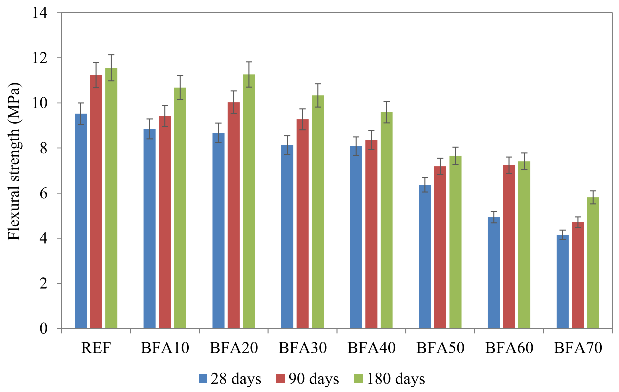

Fig. 11. Development of flexural strength after 28, 90 and 180 days of curing.

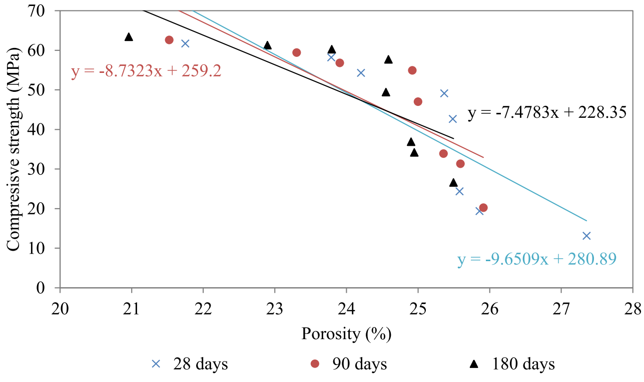

Fig. 12. Relationship between porosity and compressive strength over time.

The pozzolanic activity indices calculated on the basis of measured mechanical parameters are given in Figs. 13, 14. Considering the SAI, the BFA10, BFA20, and BFA30 mixtures exceeded the threshold values of 0.75 after 28 days and 0.85 after 90 days. Therefore, they could be accepted as structural materials. The data after 180 days provided in Fig. 13 for a deeper understanding of SAI development over time showed a further SAI increase. PEC as a parameter suitable for a more detailed assessment of BFA pozzolanic activity showed that after 28 days of curing BFA10, BFA20, BFA30, and BFA40 mixtures could be considered as pozzolan active, while the rest remained unreactive. Apparently, in the mixtures with higher BFA content, this admixture acted mainly as a filler. Nonetheless, contrary to the results achieved for 28 days, the data for 90 and 180 days of curing indicated some pozzolanic effectiveness even for BFA50, BFA60, and BFA70. This finding was in a good agreement with Demis et al. [24] where retardation of BFA incorporation into the matrix was found typical for wood-based fly ash.

It should be noted that SAI and PEC are not the only criteria that can be used for the assessment of the suitability of biomass ashes in cementitious mixtures. As mentioned by Farinha et al. [25], there are many other variables that should be taken into account in that respect, among them the amount of reactive compounds and the particle size [1,21,44]. The utilization of advanced design tools [34] may also contribute to more favorable results.

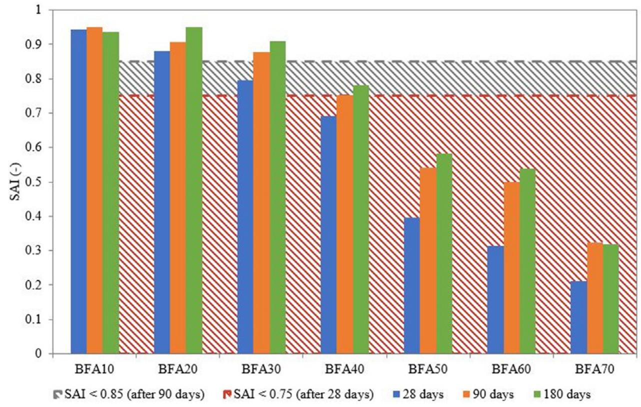

Fig. 13. SAI values of studied mortars after 28, 90 and 180 days.

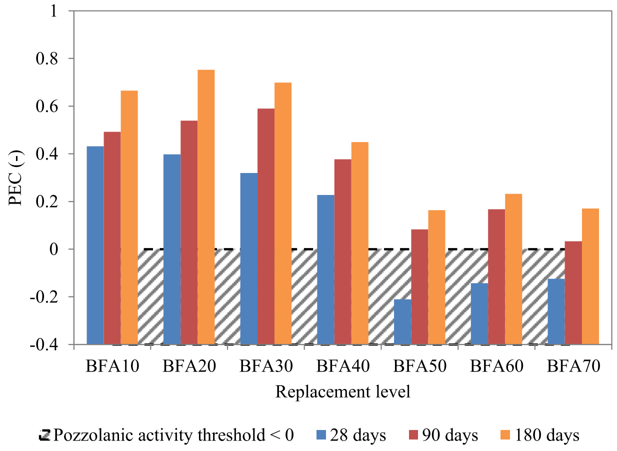

Fig. 14. PEC values of studied mortars after 28, 90 and 180 days.

5. Conclusions

As the traditional coal combustion as one of the main energy sources may be gradually abandoned in the EU in future decades, the availability of the widely applied coal fly ash (CFA) can decrease substantially compared to the present and its replacement by other available and suitable SCMs in blended cements may become a necessity. In this paper, the application of biomass fly ash (BFA) as a possible alternative to CFA was analyzed. In the experimental research, mortars containing up to 70 wt% of BFA as Portland cement replacement were investigated to determine the influence of BFA on functional properties. The following conclusions can be drawn:

- A major advantage of the studied BFA compared to CFA consisted predominantly of a substantially lower concentration of hazardous elements. Ordinary CFA obtained from coal combustion requires additional processing to mitigate the environmental hazards associated with its application. On the contrary, the analyzed BFA did not require any further processing, had a similar particle size distribution to Portland cement and pozzolanic activity of 606 mg CaO/g, which made good prerequisites for its use in blended cements.
- The isothermal calorimetry showed that the addition of BFA decreased the hydration heat evolution which presented an advantage; the high-temperature gradients in cement composites can be accompanied with thermal contraction and crack formation.
- The compressive strength after 28 days of curing gradually decreased with the increasing BFA dosage, which was in accordance with the increased porosity. On the other hand, the porosity of BFA containing mortars decreased after 90 and 180 days due to the pozzolanic reaction. The densification of mortars with higher BFA content resulted in a substantial improvement of compressive strength. Flexural strength exhibited lower sensitivity to the amount of used BFA than compressive strength. In summary, the application of up to 30 wt% of BFA as Portland cement replacement resulted in only minor alteration of functional properties.
- The pozzolanic activity tests using the SAI and PEC indices showed that after 28 days of curing the mixtures containing up to 30 wt% (SAI) or even 40 wt% (PEC) of BFA could be considered as pozzolan active, while the rest remained unreactive. Nonetheless, contrary to the results achieved for 28 days, the PEC data for 90 and 180 days of curing indicated some pozzolanic activity even for mortars with the BFA content of 50–70 wt%.

CRediT authorship contribution statement

Jan Fořt: Conceptualization, Visualization, Methodology, Funding acquisition, Supervision, Writing - original draft. **Jiří Šál:** Investigation, Data curation. **Radek Ševčík:** Visualization, Investigation, Writing - original draft. **Magdaléna Doleželová:** Investigation, Data curation. **Martin Keppert:** Investigation, Data curation. **Miloš Jerman:** Investigation, Data curation. **Martina Záleská:** Investigation, Data curation. **Vojtěch Stehel:** Funding acquisition, Validation. **Robert Černý:** Supervision, Conceptualization, Methodology, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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3.2 Replacement of cement through alkali-activation of waste brick powder

Selected journal paper

Fort, J., Vejmelkova, E., Konakova, D., Alblova, N., Cachova, M., Keppert, M., Rovnanikova, P., Cerny, R., 2018. Application of waste brick powder in alkali-activated aluminosilicates: Functional and environmental aspects. *Journal of Cleaner Production 194, 714-725*

This paper contemplates the complete replacement of cement in construction materials by alkali-activation of waste products, brick dust in particular. Contrary to the majority of papers aimed at highly amorphous precursors, the brick powder originating during brick block grinding was used. Moreover, the use of this material refers to the potential utilization of end-of-life buildings built from bricks, and the consequent rationalization of the bulk materials streams between the demolition sites and the production of new materials. Within this section, the integration of functional and environmental parameters was introduced as an advanced tool for the assessment of building materials.

Contribution to the practice: *The main findings of this paper applicable to building practice can be found in the possible utilization of end-of-life buildings from red bricks as potential resources bank for the recycling and consequent manufacturing of new construction materials.*

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Application of waste brick powder in alkali activated aluminosilicates: Functional and environmental aspects

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ABSTRACT

Several alkali activated aluminosilicate (AAA) materials prepared using waste brick powder as precursor are analyzed from both functional and environmental points of view. The functional properties in hardened state are assessed by the measurement of basic physical properties and mechanical parameters. The environmental impacts are examined by determination of energy consumption and carbon dioxide emission related to their production. The obtained results show that functional properties of designed AAAs are comparable with cement-based materials. Their environmental qualities are though substantially higher, which is due to lower demand for common raw materials and lower greenhouse gases emissions and energy consumption. The combined assessment of functional and environmental aspects, which is performed using the energy consumption efficiency index and the carbon dioxide emission efficiency index, reveals that the analyzed AAA mixes can provide up to 45% savings in consumed energy and even 72% in emitted greenhouse gases, as compared with Portland cement paste.

1. Introduction

Since cement manufacturing was recognized as highly energy demanding process with significant negative environmental consequences, the urge to improve environmental sustainability of building materials became an important issue. The production of Portland cement as the most frequently used binder is responsible for almost 5% of world carbon dioxide emissions (IEA, 2008). According to other studies (Allwood et al., 2010), its share can be even higher, 6–8%. Therefore, an extensive research has been aimed at the strategies reducing CO_2 emissions within the past decades. The effort was concentrated mainly on the following areas: substitution of clinker by mineral admixtures, increasing energy efficiency and use of alternative fuels (Summerbell et al., 2016). Besides these main strategies, the use of new binders attracted a considerable attention. Alkali activated materials represented an alternative and less energy demanding solution than ordinary Portland cement based composites (Provis et al., 2015). Alkali activated aluminosilicates (often denoted also as geopolymers) exhibited, according to the previous studies, promising and excellent physical properties, such as durability, acid and sulfate resistance and high temperature resistance, which can be utilized in various applications (Gluknovsky, 1959; (Duxson et al., 2007); (Pan et al., 2018); (Hossain et al., 2018)).

The first experience with alkali activated materials is dated to the ancient times but their first applications in the modern era started in 1940s (Pacheco-Torgal et al., 2008). In 1980s, mixing of alkalis with fine particles of natural minerals became more frequent and new binders called geopolymers were introduced by Davidovits (1998), who invented many different solutions and indicated their consequent possible applications in building industry. Nowadays, geopolymers are subject of intensive investigations by various research groups. A representative state of the art can be found in the extensive report edited by Provis and van Deventer (2014).

From a theoretical point of view, any building material with silica and alumina content can be utilized as alkali activated material. The use of supplementary cementitious materials in building industry represents a promising way to reach sustainable solutions while preserving necessary functional properties. The

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investigations of prospective materials were aimed mainly at metakaolin (Alonso and Palomo, 2001; Bouguermouh et al., 2017), kaolinic clays (Barbosa et al., 2000; Longhi et al., 2016), blast furnace slag (Hwang et al., 2017; Wang et al., 2005), mixtures of slag and various additives, and fly ashes (Samson et al., 2017; Xu et al., 2017) to date. Utilization of industrial by-products or waste products is commonly considered very beneficial for the development of binders with lower environmental impact.

Ceramic waste belongs to materials whose alternative use can reduce the current intensive worldwide landfilling. Despite of the energy demanding requirements connected with crushing raw ceramic waste to sufficient fineness, the utilization of finely ground ceramic waste represents a promising way towards the decrease of both CO_2 emissions and costs, which are associated with extensive applications of ordinary Portland cement (Robayo-Salazar et al., 2017). Compared to Portland cement, ceramic waste does not require any further processing, except for grinding. It can be used in the form of aggregates in cement based composites (Lucas et al., 2016) or as a partial replacement of Portland cement (Puertas et al., 2008; Vejmelkova et al., 2012). Some applications of waste ceramic materials for the preparation of alkali activated aluminosilicates were reported as well (Reig et al., 2013a, 2013b) but they were rare until now.

The mixtures of sodium and potassium hydroxide with sodium silicate represent the most frequently used alkaline activators for the preparation of geopolymers (Duxson et al., 2007); Provis and van Deventer, 2014). (Komljenovic et al., 2010) concluded that the most dominant factor during the alkali activation process is the type, ratio, and concentration of the used alkaline activator. The relation between concentration of alkaline activators and final mechanical properties was also reported (Fernandez-Jimenez and Palomo, 2005); (Wang et al., 2005), where higher molar concentrations led to better mechanical properties. (Ken et al., 2015) found, based on the work focused on the combination of alkaline activators, pure water glass or its combination with sodium hydroxide as the most promising way for obtaining the best compressive strength.

Thanks to the minimal requirements of geopolymers on processing and depletion of natural resources, the reduction of human environmental footprint presents an essential factor for their wider application. Surprisingly, opposed to the initial assumptions considering different processing of source materials necessary for the production of geopolymers, contradictory conclusions can be found in literature in that respect. On one hand, Habert et al. (2011) or (Turner and Collins, 2013) rejected geopolymers as an environmental friendly solution due to a significant negative influence of other impact categories, such as abiotic depletion, acidification, eutrophication, and human toxicity, which were increased particularly due to the amount of used alkaline activators. Negative implications of utilization of alkali activated materials were discussed also by Davidovits (2005). On the other hand, several studies (Davidovits, 1998; Komnitsas and Zaharaki, 2007; Van Deventer et al., 2012) posed alkali activated materials as a prospective and favorable solution for a reduction of consumed energy and emitted greenhouse gases (GHGs). Stengel et al. (2009) and Weil et al. (2009) in more specific investigations reported 45% and 70% savings in these two impact categories, respectively. (McLellan et al., 2011) also calculated possible cost savings compared to the application of Portland cement, even though these data can be a subject of further discussion due to the Australia local specifics, such as long transportations distances.

It should be noted that many studies focused on the evaluation of environmental impact of alkali activated aluminosilicates were not accompanied with the assessment of functional properties of studied materials. Conversely, many investigations dealing with the influence of various activators and resulting mechanical properties disregarded any environmental assessment of studied materials. Hence, despite the significant increase of the number of studies focused on the utilization of waste or by-products in alkali activated binders, the developed products mostly require extensive investigations to deliver more representative information on their overall quality. The great potential of alkali activated binders to decrease negative externalities connected with the processing of conventional building materials, together with all demanding requirements and problems associated with their production and applications, presents good opportunities for material engineers to develop novel materials with unusual combinations of functional and environmental properties.

In this paper, several alkali activated aluminosilicates (AAA) are designed, using waste brick powder as precursor and a mix of water glass and sodium hydroxide as alkaline activator. The main goal of this study is to provide a complex assessment of the analyzed materials, taking into account and balancing both functional properties in hardened state and environmental impacts.

2. Materials and samples

2.1. Applied materials

A waste brick powder generated by grinding of thermal insulation bricks was used for the development of the alkali activated binders. The additional advantage of this material, besides being a waste product, laid in its ready-to-use properties, which did not require any thermal treatment or crushing. The collected material was dried at 105 °C for 48 h to remove redundant moisture. Then, the dried brick powder was sieved, in order to separate larger particles or shards and obtain a homogenous material. Only particles smaller than 0.125 mm were alkali activated because of their better reactivity promoting mechanical performance of designed materials in hardened state (He et al., 2013).

For the alkali activation, a mixture of sodium hydroxide pellets (99%, Fichema a.s., Czech Republic) and sodium silicate (water glass) with SiO_2/Na_2O molar ratio of 1.6 (Vodní sklo, a.s., Czech Republic) was used.

2.2. Sample preparation

At the preparation of mixtures of alkali activated materials, sodium hydroxide solid pellets were dissolved in water at first. Then, water glass was added to the solution in the predetermined ratio. The obtained alkali activator was mixed together with the desired amount of waste brick powder. The AAA pastes were prepared at atmospheric pressure by mixing the activator with water and brick powder. Five different AAA mixtures were prepared, denoted as AAP 60, 70, 80, 90 and 100 according to the amount of used water glass. The composition of studied materials is summarized in Table 1.

Samples were cast into 40 mm × 40 mm x 160 mm molds and kept in a climatic chamber at 20 °C and 50% relative humidity for 7 days, taking into account their possible longer plasticity time period (Messina et al., 2018). After demolding, samples were placed again in a climatic chamber at the same conditions for next 21 days. All materials were tested after finishing 28 days curing at described conditions.

3. Assessment methods

3.1. Material characterization

An Analysette 22 Micro Tec plus (Fritsch) laser diffraction device

Table 1
Composition of the studied mixtures.

	Waste brick powder (g)	Water glass (g)	NaOH (g)	Water (ml)	Silicate modulus of the activator (–)
AAP60	200	60	2.19	46	1.4
AAP70	200	70	2.57	41	1.4
AAP80	200	80	2.95	36	1.4
AAP90	200	90	3.33	31	1.4
AAP100	200	100	3.76	26	1.4

with the measuring range up to 2 mm was used for determination of the particle size distribution of the applied waste brick powder. Large particles were detected using an infrared laser with a large distance to the measuring cell, for small particles a green laser with a small distance to the cell was used, which permitted detection of forward scattered light up to a scattering angle of 65°. The measurement of smallest particles down to the nano-range was performed by the green laser light for backward scattering.

The phase composition was examined by X-ray diffraction (PANalytical X'PertPRO diffractometer equipped by CoKα X-ray tube (40 kV, 30 mA)). The data were evaluated by the HighScorePlus software package (version 3.0.5) and JCPDS PDF2 database. The Rietveld analysis was performed using Topas software to determine the content of crystalline phases. The samples for XRD analysis were pulverized using agate mortar to pass 20 μm sieve. The quantification of amorphous portion was performed by help of internal standard (ZnO, 10%). The chemical composition was determined by XRF spectroscopy (Thermo ARL 9400 XP). The reproducibility of the measurement was 0.0001% and the standard measurement error 0.02%.

3.2. Functional properties

Among the basic physical properties of studied AAA materials, the bulk density, matrix density, and open porosity were measured. The bulk density was determined by weighting of the samples with known volume which was obtained using a digital caliper. The matrix density was measured by a helium pycnometer Pycnomatic ATC (Thermo Scientific). The total open porosity ψ was calculated on the basis of the knowledge of bulk- and matrix density according to the following equation:

$$\psi = 100\left(1 - \frac{\rho_b}{\rho_{mat}}\right) \tag{1}$$

where ρ_b (kg/m^3) is the bulk density and ρ_{mat} (kg/m^3) the matrix density of the material.

The mercury intrusion porosimetry (MIP) analysis was done to characterize the inner structure of studied materials. Pore size distribution was measured using a combination of porosimeters Pascal 140 and Pascal 440 (Thermo Scientific). At the evaluation of measured data, the circular cross section of capillaries was assumed, whereas the mercury contact angle was assumed to be 130°.

A hydraulic testing device VEB WPM Leipzig having a stiff loading frame with the capacity of 3000 kN was employed for the measurement of compressive and bending strength. The flexural strength was determined using the procedure described in ČSN EN 12390-5. For the measurement, prismatic samples having dimensions of 40 mm × 40 mm x 160 mm were used. The compressive strength was measured according to ČSN EN 12390-3 on the portions of prisms broken in the bending test; the loading area was 40 mm × 40 mm.

3.3. Environmental impact

Taking into account the contradictory statements on the environmental impact of AAA found in the literature, which were mentioned in the Introduction section, a simplified environmental analysis was done to quantify and compare environmental impacts of the application of waste brick powder in alkali activated materials.

3.3.1. Goal and scope

In order to access the environmental impact of the alkali activated aluminosilicates based on waste brick powder, two basic comparative criteria were chosen: the amount of consumed energy and carbon footprint.

The range of the environmental analysis was reduced to the intermediate stage of material processing (cradle to gate), while the service life of the material in a structure was neglected due to a lack of information about the material long-term durability. This approach can be applied also because of the possible multiple specific applications in civil engineering with different life cycles. The reason for the chosen system boundaries lies in the effort to maintain quality and reproducibility of the used analysis. The systems boundaries showed in Fig. 1 secure the framework of the comparison and collection of energy and material inputs and outputs created during material processing.

For the fulfillment of requirements related to the comparative assessment of particular mixtures, a functional unit of 1 ton of final material was used. In order to evaluate the potential environmental benefits of the studied alkali activated aluminosilicates, their environmental impacts were compared with Portland cement paste (PCP).

3.3.2. Inventory analysis

The emission data and consumed energy necessary for the Life Cycle Inventory were compiled from the literature review, theoretical estimations and information provided by the producers. Firstly, relevant manufacturing processes were identified (Fig. 2). The emphasis was placed on the consideration of local factors, such as composition of energy mix or variants of the used materials. Within the analysis, the Czech electricity mix, where the main portion of electricity comes from coal combustion and nuclear power plants was considered (OTE, 2016). The energy data sources obtained by literature review are compiled in Table 2, where some specifications and range of analysis are noted. The emissions related to potential transportation are neglected due to the incomparability with the current cement industry. The justification of the exclusion of these expenditures lays in the good establishment of cement industry, where almost all components are manufactured on the same place in order to increase effectiveness of the whole processing and decrease the costs, contrary to the new binder which logically did not achieve yet this level of optimization.

3.3.3. Impact assessment

The impact categories of cumulated energy consumption and

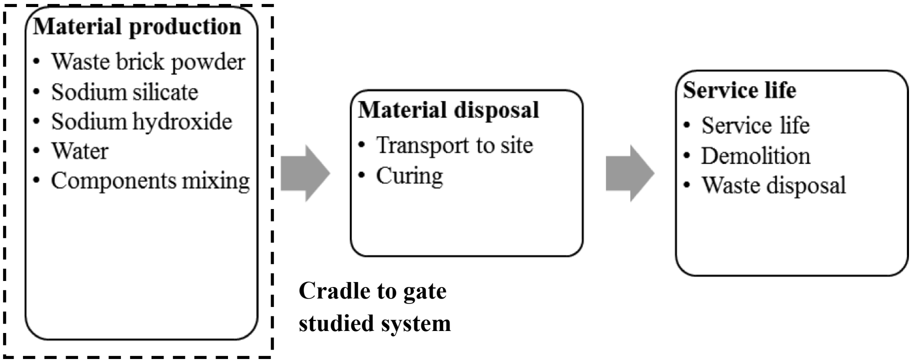

Fig. 1. Boundaries of environmental analysis.

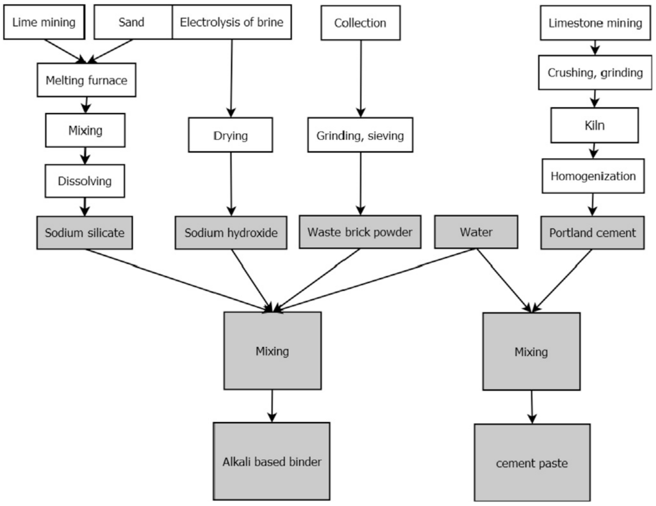

Fig. 2. Identification of relevant processes related to the material production.

greenhouse gases (GHG) emissions were taken into account during the life cycle assessment. The GHGs emissions were stated in kg CO_2 equivalent and the particular importance of gases with the most harmful effect on global warming were included. For data mining, it was important to deliver proper data related to Europe and respecting the system boundaries to provide correct and comparable analysis, as neglecting the regional influences can lead to serious misinterpretations.

The applied brick powder as a waste material being currently landfilled does not need any substantial manufacturing process, except for sieving and drying. This advantage gives it a considerable potential for utilization in the design of materials with a lowered impact on the environment. This fact is justified by the assumption that consumed energy and generated emissions are related to the processing of the main product. Hence, all negative externalities are connected with the processing only; wastes or by-products are free of all embodied impacts.

3.4. *Combination of functional and environmental aspects*

In order to provide a complex view of the potential of the designed alkali activated binders in the building industry, a combined assessment of the functional and environmental aspects was done using the binder use efficiency indicators. As the supposed applications of alkali activated binders are similar to cement, the indicators proposed by (Damineli et al., 2010) to estimate efficiency of cement use were adopted in this paper.

The CO_2 intensity index C, which presents a modification of the original f_{eco} index introduced by (Popovics, 1990), comprises the environmental loads in terms of CO_2 emissions imposed to deliver a unit of functional performance measured by a suitable indicator (compressive strength, bending strength, etc.). It is defined as:

$$C = \frac{c}{p} \tag{2}$$

where c (kg CO_2) is the total CO_2 emission related to the functional unit and as p the compressive strength R_c (MPa) is used in most cases.

The energy consumption efficiency index E, which provides a transparent comparison of the expended energy per a unit of functional performance, is utilized as the second assessment criterion,

$$E = \frac{e}{p} \tag{3}$$

where e (MJ) is the energy related to the production of a functional unit of the binder and p is a unit of functional performance (mostly compressive strength, R_c).

4. Results and discussion

4.1. *Material characterization*

The particle size distribution of the applied waste brick powder is presented in Fig. 3. The obtained data revealed a main peak at

Table 2
Emission data used in the environmental analysis.

Material	Specification	Analysis range	Key references
Portland cement	product	mining, grinding, calcination, storage	(Chen et al., 2010; Damineli et al., 2010; Feiz et al., 2015; Garcia-Gusano et al., 2015; Giama and Papadopoulos, 2015; Huntzinger and Eatmon, 2009; Josa et al., 2007).
Sodium silicate	product	mining, melting, dissolving	Fawer et al., 1999; Althaus et al., 2007; Zoller and Sosis, 2008.
Sodium hydroxide	product	electrolysis, drying	Wilson and Jones, 1994; Althaus et al., 2007; Thannimalay et al., 2013.
Waste brick powder	waste product	collection	Puertas et al., 2008; Reig et al., 2013a; Kočí et al., 2016.

61 μm, which could be considered as a satisfactory fineness; the brick powder was only somewhat coarser than Portland cement. The importance of the particle size distribution was observed by several authors who found its impact on final mechanical properties (He et al., 2013; Sate et al., 2012). Finer particles of precursor were reported to have a better reactivity and be able to form denser geopolymer structure (Nazari et al., 2011).

The X-ray fluorescence (XRF) analysis of chemical composition of the studied waste brick powder (Table 3) showed that its major part was formed by SiO_2 (58.8%), while Al_2O_3 constituted 19.6%. The obtained composition was closer to the mixture of red mud and metakaolin used by (Ye et al., 2016) than to traditionally used calcium rich precursors (Chotetanorm et al., 2013). The reactivity of ceramic waste powders is, according to (Provis et al., 2015), lowered due to the higher burning temperature compared to metakaolin. The conceptual model based on such materials was thus not suitable for a description of alkaline activation of waste brick powder used in this paper.

The mineralogical composition obtained by XRD (Table 4) was similar to the red clay waste brick used by (Robayo-Salazar et al., 2017), who though did not estimate the amorphous content. In this paper, the amorphous content of the studied waste brick powder was only 27.8 wt% (Table 4), which could present a limiting factor, as for the precursor reactivity (Pacheco-Torgal et al., 2008; Reddy et al., 2016).

As the reactivity of the analyzed brick powder is supposed to depend significantly on the content of amorphous silica and alumina, the chemical composition of the amorphous phase was estimated using the data in Tables 3 and 4. The results are presented in Table 5. Apparently, the amounts of SiO_2 and Al_2O_3 were very different from the whole composition data.

Davidovits (1999) suggested, based on the zeolite chemistry, that SiO_2/Al_2O_3 molar ratios can be considered as indicators for a preliminary estimation of mechanical properties. He proposed the range of 2–5.5 to deliver the best mechanical performance. Similarly, Provis and van Deventer (2014) and (Autef et al., 2016) considered the optimal SiO_2/Al_2O_3 molar ratio to be higher than 2.5. However, some other authors considered this statement as arguable because of differences in composition of various precursors (Chen et al., 2017; Yuan et al., 2016) and the different reactivity of silica and alumina (Criado et al., 2008; Pacheco-Torgal et al., 2008; Ruiz-Santaquiteria et al., 2013). In the comprehensive study by (Xu and Van Deventer, 2000) the prediction of suitability of materials for geopolymerisation based only on the molar SiO_2/Al_2O_3 ratio was dismissed due to the extreme complexity of gel formation and hardening mechanisms. Therefore, one can conclude that the opinions of various researchers on the importance of the SiO_2/Al_2O_3 ratio differ each other and a coherent theory in that respect was not formulated yet. It should also be noted that most researches were performed for materials with dominant amorphous part to date. In case of precursors with minor amorphous parts it is important to distinguish between the individual phases (crystalline and amorphous) and molar ratios should be calculated for both (Sassoni

Table 3
Chemical composition of waste brick powder (XRF).

Compound	SiO_2	Al_2O_3	Fe_2O_3	CaO	K_2O	MgO	Na_2O	TiO_2	SO_3
(% by mass)	58.8	19.6	5.7	6.9	2.9	2.8	1.5	0.8	0.7

Table 4
Mineralogical composition of waste brick powder (XRD).

Substance	Chemical composition	(% by mass)
Amorphous part	–	27.8
Quartz	SiO_2	26.2
Hematite	Fe_2O_3	2.3
Albite	$NaAlSi_3O_8$	13.0
Microcline	$KAlSi_3O_8$	3.6
Orthoclase	$KAlSi_3O_8$	3.5
Muscovite	$KAl_2(AlSi_3O_{10})(OH)_2$	12.5
Illite	$K_{0.65}Al_2(Al_{0.65}Si_{3.35}O_{10})(OH)_2$	3.8
Diopside	$CaMgSi_2O_6$	4.4
Akermanite	$Ca_2MgSi_2O_7$	2.8

Table 5
Chemical composition of the amorphous phase of waste brick powder (XRD).

Compound	SiO_2	Al_2O_3	Fe_2O_3	CaO	K_2O	MgO	Na_2O	TiO_2	SO_3
(% by mass)	27.6	34.5	12.2	16.6	0.0	5.6	0.0	2.9	2.5

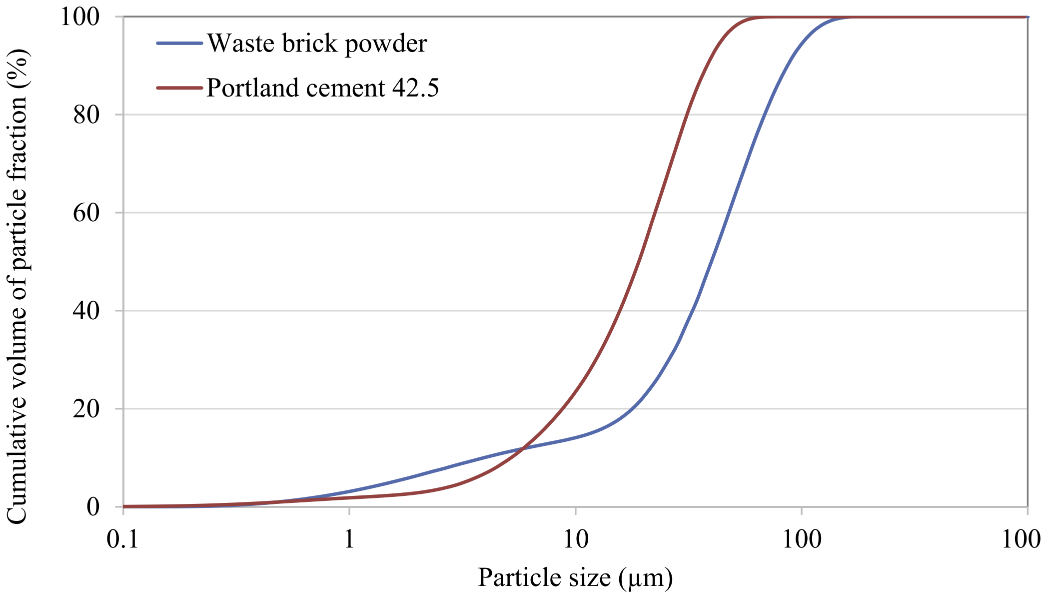

Fig. 3. Particle size distribution curve of waste brick powder and Portland cement.

et al., 2016). In this paper, the total SiO_2/Al_2O_3 molar ratio of the waste brick powder was 1.73 (Table 3), while the SiO_2/Al_2O_3 molar ratio of the amorphous part was 0.49 (Table 5).

4.2. *Functional assessment*

The basic physical properties of studied alkali activated materials are presented in Table 6. Mixtures denoted as AAP70-100 formed a dense and compact structure while AAP60 was not able to form a compact material and remained plastic even after a long curing time. This finding can be assigned to the poor ability of applied alkali activators to break covalent Si—O—Si and Al—O—Si bonds due to the low amount of used NaOH (Xie and Xi, 2001). The addition of higher amounts of NaOH significantly improved the ability of the mixtures to create a condensed structure. A higher dosage of alkaline activators slightly increased the bulk density while the matrix density retained almost the same values around 2625 kg/m^3. The total open porosity of about 35% was similar for all mixtures, except for AAP100 with 38.5% which correlated with the lower bulk density. The drop in bulk density observed for AAP100 mixture can be assigned to the changes in hydration kinetics and rheology of the fresh mixture. While other mixtures remained plastic for certain time period, the AAP100 mixture exhibited low plasticity. Therefore, mixing and material compacting was limited. The change of pore structure was also promoted by the different SiO_2/Al_2O_3 molar ratio which could lead to a decrease of strength (Sassoni et al., 2016; Guo et al., 2017).

A more detailed insight into the pore characterization is given in Fig. 4, where the cumulative pore size distribution curves determined by MIP are plotted. Here, the AAP60 mixture could not be included again for its hardening problems. The other mixtures exhibited significant differences. The AAP70 mix showed the lowest total pore volume in the range of 1–100 μm. This finding was related to its more favorable SiO_2/Al_2O_3 molar ratio which allowed for a good level of geopolymerization (Fletcher et al., 2005; Reddy et al., 2016). The differences in the cumulative pore volume between AAP70 and the remaining materials were significantly higher than differences in the total open porosity determined for bigger samples in Table 6. The observed pore size distribution changes were also reflected in the changes of average pore diameter which was significantly lower for AAP70. This fact indicated the presence of a substantial amount of pores larger than 100 μm in AAP80, AAP90, and AAP100 in particular. The differences in pore structure between AAP70 and other mixtures were related, apparently, to the different dosage of the used alkaline activator; the increase of its amount in the mix led to an increase of SiO_2/Al_2O_3 molar ratio (Komnitsas et al., 2015).

The compressive and bending strength of the analyzed AAA pastes based on waste brick powder are presented in Fig. 5. Here, the obtained results are plotted together with the mechanical properties of PCP to show a clear comparison since geopolymers present a supposed alternative to cement based materials. The mechanical performance of cement paste can be perceived as reference for that reason, despite of the different hydration process and formed products. The results for AAP60 were not included because the material did not achieve hardened state and remained plastic even after a long curing period. Looking at the plotted results, the best performance was obtained for AAP70 mixture with the compressive strength, R_c, of 41.9 MPa, while AAP80, AAP90, and AAP100 achieved only $R_c = 36.5$ MPa, 29.1 MPa, and 10.3 MPa, respectively. The most distinct drop was observed for AAP100. The data obtained for bending strengths followed a similar trend. The AAP70 mixture with 11.9 MPa achieved even higher bending strength than PCP, which could be considered as a very good result.

Table 6
Basic physical properties of studied materials.

Material	Bulk density (kg m^{-3})	Matrix density (kg m^{-3})	Open porosity (%)
PCP	1679	2208	24.0
AAP60	–	–	–
AAP70	1697	2637	35.5
AAP80	1711	2626	34.8
AAP90	1719	2622	34.4
AAP100	1611	2620	38.5

A comparison of mechanical parameters of alkali activated waste brick powder measured in this paper with the results obtained by other investigators could be done in a limited extent only, due to the differences in chemical composition and SiO_2/Al_2O_3 molar ratio of applied precursors. The achieved results represent significant improvements in mechanical properties, compared to (Pathak et al., 2014). On the other hand, (Robayo-Salazar et al., 2017) reached significantly higher values of compressive strength (up to 102 MPa) by alkaline activation of red clay waste brick but the results were affected by the addition of Portland cement. Better comparable studies were carried out by (Reig et al., 2013a, 2013b) who used construction and demolition waste for alkaline activation and reached compressive strengths of 22–41 MPa after 7 days of temperature curing at 65 °C. The authors further improved mechanical properties by optimization of the used alkaline activator and water/binder ratio up to 50 MPa. (Sun et al., 2013) achieved for alkali activated ceramics the compressive strength of 71 MPa by curing at 60 °C for 27 days. Despite of the obvious importance of bending strength, many authors avoided determination of this parameter and remained focused on the compressive strength only. Bernal et al. (2012), who studied mechanical properties of geopolymers based on blast furnace slag and metakaolin, belonged to the few exceptions in that respect. They showed that the prolongation of curing period can significantly increase bending strength; the highest value of about 10 MPa was achieved after 90 days of curing. In this paper, for the AAP70 mixture the bending strength of 11.9 MPa was obtained after 28 days of curing and despite of the lower amorphous content of waste brick powder in a comparison with metakaolin or blast furnace slag. Lo et al. (2018) observed that bending strength was substantially affected by the degree of polymerization and optimized the SiO_2/Na_2O ratio to get a maximum value of 10.4 MPa, which was also somewhat lower than bending strength of the best mix in this study.

On the other hand, the properties of PCP which can serve best for a complex comparison with the AAA designed in this paper from functional and environmental points of view, were published frequently. For instance, (Su et al., 2015) studied the effect of various w/c ratios, as well as the influence of the size effect on the cubic compressive strength of cement paste. Here, the compressive strength varied from 22.1 MPa in the case of w/c = 0.6–66.5 MPa for highly optimized cement pastes with w/c = 0.2. Considering the results obtained in this paper for AAA based on waste brick powder from this angle, the values of compressive strength after 28 days of curing (Fig. 5) were satisfactory and competitive with cement pastes.

The observed decrease of the values of mechanical parameters of the analyzed AAA with an increasing amount of used alkaline activators was related, apparently, to the changes in microstructure of the studied materials. It correlated well with the presence of a substantial number of pores larger than 100 μm in AAP80, AAP90, and AAP100 in particular, which was indicated by the discrepancies between the porosity values measured by MIP and calculated from the values of bulk density and matrix density (Table 6 and Fig. 4). The finer and more homogeneous microstructure of AAP70, as compared with AAP80, AAP90, and AAP100, was thus the most

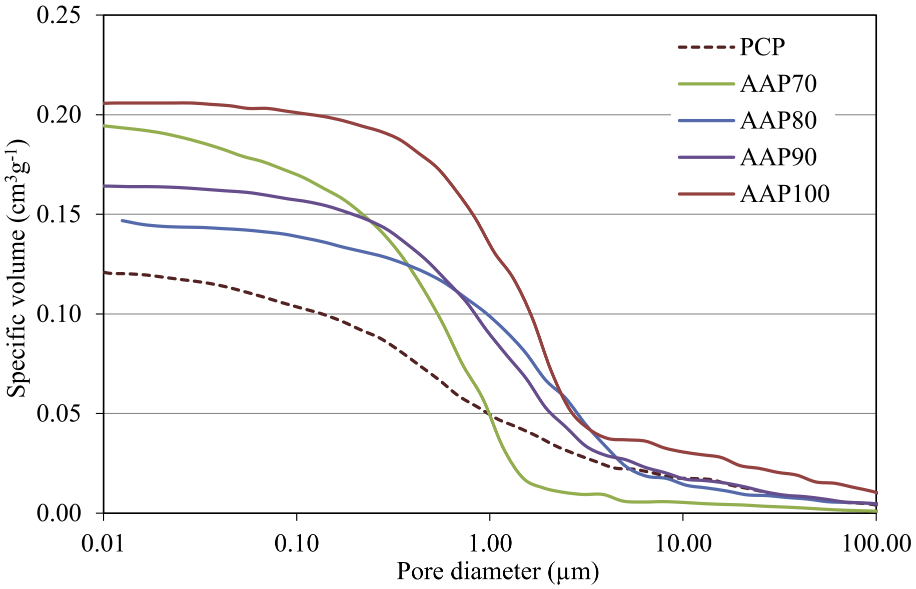

Fig. 4. Pore size distribution of studied AAA pastes.

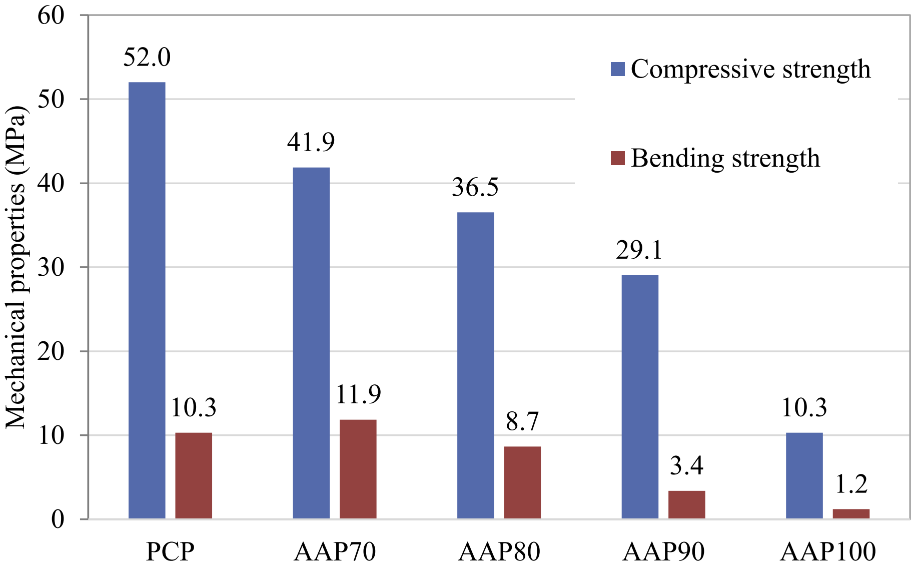

Fig. 5. Mechanical properties of studied AAA pastes after 28 days.

important factor affecting the mechanical properties of the analyzed AAA. The total open porosity, on the other hand, showed an opposite trend than for most cement based materials; it decreased with the decreasing strength values and could not be considered as a reliable strength indicator, except for the AAP100 mixture. Similar findings were reported by (Komnitsas and Zaharaki, 2007; Somna et al., 2011), who showed that an increase in concentration of applied alkaline activators induced structural changes due to undesirable molar ratios during the hydration period which resulted in a decrease of compressive strength.

The mechanical parameters of AAA certainly depend on the chemical and physical nature of the particular phases formed in the geopolymerization process. However, these phases are studied only rarely in detail, mainly due to the questionable determination of the exact boundary between the crystalline and amorphous phase of the material, as it was pointed out by Provis et al. (2005). In this paper, a basic characterization of hardened AAA pastes was carried out using the XRD analysis aimed at the quantification of the amorphous phase. An example of the obtained X-ray diffractograms is given in Fig. 6 for AAP70. Here, quartz (SiO_2), muscovite ($KAl_2(AlSi_3O_{10})(OH)_2$) and albite ($NaAlSi_3O_8$) were identified as the most distinct crystalline phases which though were most important also in the crystalline part of the brick powder itself (cf. Table 4). Therefore, they presented the non-reacted components of the brick powder. Quartz, muscovite, or albite as the most distinct phases in the raw waste brick powder were reported in final geopolymer products by other researchers (Komnitsas et al., 2015; Reig et al., 2016; Sun et al., 2013) as well, pointing at a lower (if any) reactivity of crystalline phases. On the other hand, zeolitic phases identified in alkali activated pastes by (Bernal et al., 2010) or (Guo et al., 2010), were not found in the studied AAA. The content of amorphous phase was in all analyzed materials higher than in the precursor, 40.1% by mass in AAP70, 45.1% in AAP80, 44.7% in AAP90, and 43.8% in AAP100. The reaction products of AAA70 with the

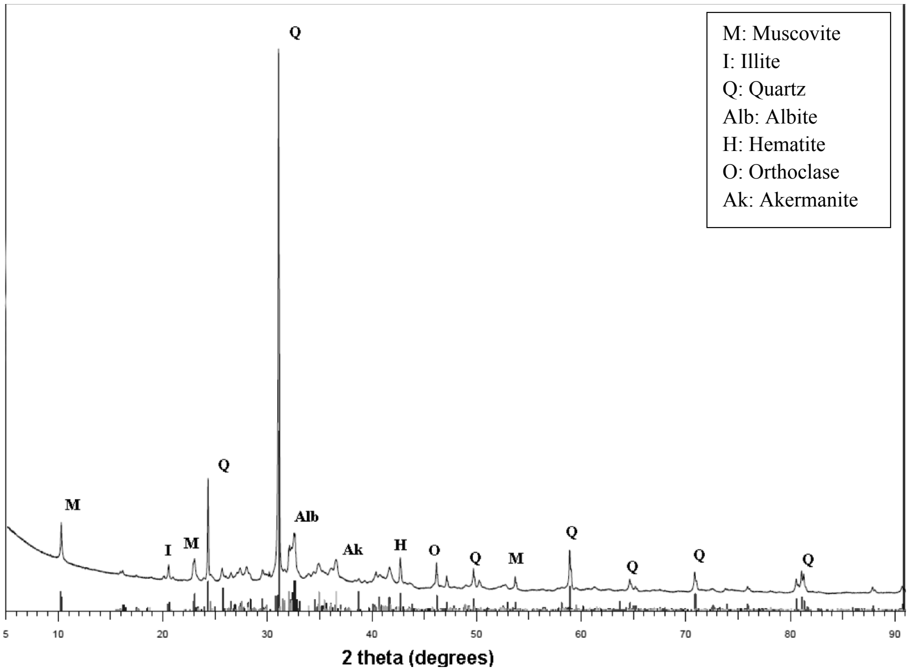

Fig. 6. X-ray diffractogram of AAP70.

lowest content of amorphous phase exhibited the finest pore structure (Fig. 4) which correlated with the highest strength of AAA70 (Fig. 5). Apparently, the crystalline phase of the precursor could reinforce the resulting consolidated materials, as it was suggested by Gharzouni et al. (2016), despite of general considerations of higher amorphous content as a factor promoting better reactivity in contact with alkaline solution (Cristelo et al., 2016; Singh and Subramaniam, 2017).

4.3. Environmental assessment

Although the development of new eco-efficient materials is a motivation widely pronounced by many authors, the assessment of environmental factors is often neglected (Ferdosian and Camoes, 2017; Medina et al., 2017; Nehdi et al., 2017). Therefore, the estimation of environmental impact of material production can be perceived as an important criterion for a complex material assessment. The particular data of energy consumption and GHGs emissions of the components of AAA mixes designed in this paper and Portland cement serving for a comparison are given in Table 7 where a substantial contribution of the alkaline activators on the results is distinct.

Figs. 7 and 8 present the related cumulative energy consumption and emission of GHGs of the analyzed AAA in a comparison with the PCP having the compressive strength of 52 MPa (Pavlik et al., 2016). The obtained values refer to a significantly lower negative environmental impact in these two chosen categories. Namely, 63% of the total consumed energy and 81% of the emitted GHGs were saved in the case of the AAP70 mixture, which provided the best mechanical properties among the studied AAA. Even higher savings could be achieved for AAP60 but this mixture was not able to reach hardened state in a sufficient time and had to be excluded from further considerations (see Section 4.2).

Table 7
Emissions and energy consumption demand for the individual components.

Constituent part	Energy consumption (MJ/t)	CO_2 emissions (kg/t)
Portland cement	2986	829
Sodium silicate - pure	5462	1514
Sodium hydroxide - pure	6965	1930
Waste brick powder	135	28
Mixing	21	12

In a comparison with the results reported by other investigators, which varied mostly from 30 to 75% of saved GHGs for different AAA mixes (Davidovits, 2005; Komnitsas and Zaharaki, 2007), the savings achieved for AAA in this paper were above the upper limit. It should be noted, however, that the differences between the results obtained in this paper and in the earlier studies could be caused by many factors which may be often difficult to compare exactly.

The variations in geopolymer mixtures, the type and amount of used alkaline activators and the chosen precursors present the factors influencing the results of performed studies in the most significant way (Weil et al., 2009). In particular, the used alkaline activator has a very important impact on the environmental assessment of geopolymers. The application of sodium hydroxide and sodium silicate results in significantly increased production of carbon dioxide. (Robayo-Salazar et al., 2017) suggested a replacement of traditional alkaline activators (sodium silicate and sodium hydroxide) by alternatives, such as sodium sulfate or sodium carbonate, to mitigate these negative externalities. However, the alkaline activation done by the mentioned alternative activators did not lead to sufficient mechanical properties. (Passuello et al., 2017) proposed the utilization of waste alkalis from industrial production as an efficient way to achieve more suitable solutions from the environmental point of view and pointed to future studies which should be aimed at the combination of durability of formed materials and low environmental impacts. They also highlighted that ready-to-use precursors are highly favorable for alkaline activation. From this perspective, the waste brick powder can be viewed as a progressive material regardless of the relatively low amorphous phase content, which can be further moderated by addition of aluminum silicate powder (Reig et al., 2017; Sun et al., 2013).

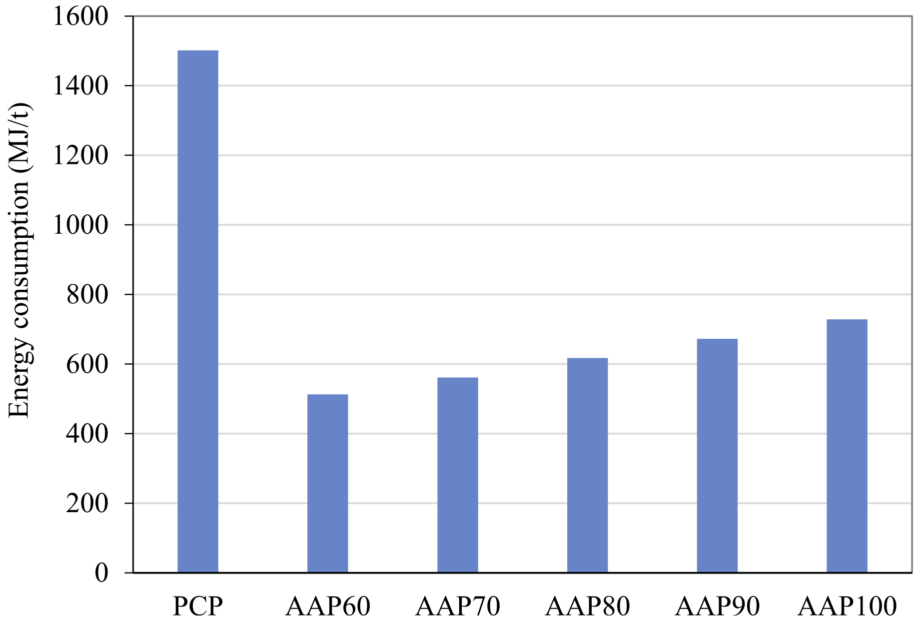

Fig. 7. Comparison of energy consumption.

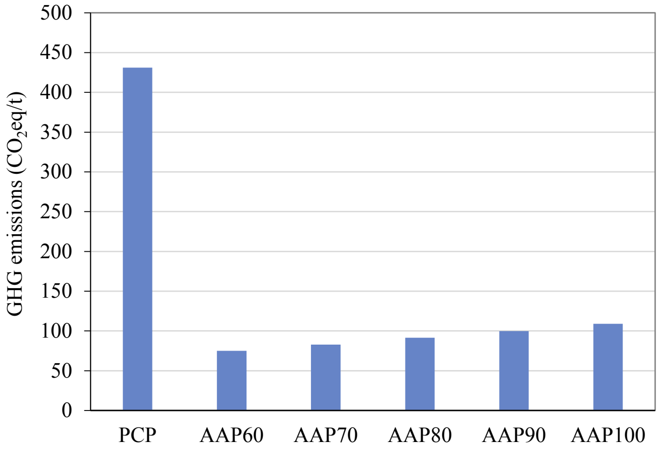

Fig. 8. Comparison of GHGs emissions.

The energy demanding process related to the production of sodium hydroxide or sodium silicate can lead to negative results, as it was reported by Habert et al. (2011). Both these components of the alkaline activator presented the highest environmental impact also in this paper. On the other hand, utilization of the waste brick powder generated by grinding of thermal insulation ceramic blocks was a very positive factor from the environmental point of view. Its application in AAA not only represented an effective solution for disposal of a waste industry product; thanks to the absence of allocation or manufacturing processes needed for its use the environmental impact of new alkali activated aluminosilicates was further decreased, as compared with most other precursors.

4.4. *Combination of functional and environmental aspects*

The materials efficiency comprising both functional and environmental properties is presented in Table 8, where the energy consumption efficiency index E and the CO_2 emission efficiency index C were calculated according to Eqs. (2) and (3). The obtained values were related to the compressive strength of 1 MPa and compared with the PCP from Pavlík et al. (2016). In light of these results it is possible to conclude that the eco-efficiency of the AAA utilization allowed a mitigation of negative environmental impacts related to the demanding Portland cement production. While in the case of PCP for the attainment of 1 MPa it was necessary to expend 31.89 MJ and emit nearly 7 kg CO_2, the utilization of AAA based on the waste brick powder led to a substantial decrease of both mentioned parameters for most analyzed AAA materials. The only exception was the AAP100 mixture where the environmental impact related to 1 MPa was higher than for the PCP, which was due to the higher amount of used alkali activator and the significant drop of compressive strength. On the other hand, the best AAP70 mixture provided savings as high as 45% in consumed energy and even 72% in the emitted GHGs. Qualitatively similar results were achieved by (Robayo-Salazar et al., 2017), who highlighted the substantial contribution of alkaline activators to environmental indicators at the development of hybrid cement containing alkali activated red clay brick waste. They achieved the most favorable results (combined functional and environmental) for the mixture with 90% of alkali activated red clay brick waste and 10% of Portland cement. Here, savings up to 80% of produced carbon dioxide were identified at material manufacturing. (Heath et al., 2014) achieved a reduction of carbon dioxide production by utilizing clay minerals and optimizing the composition of alkaline activators. However, this method was limited by the precursor specifics. Moreover, the authors used optimal molar ratios which did not distinguish between the total composition and composition of the amorphous phase.

Table 8
Combined assessment of functional and environmental properties.

Mixture	E (MJ/MPa)	C (kg CO_2/MPa)
PCP	31.89	6.97
AAP60	–	–
AAP70	13.40	1.98
AAP80	16.91	2.51
AAP90	23.15	3.44
AAP100	70.78	10.58

5. Conclusions

Waste brick powder from the production of thermal insulation bricks was used as a precursor and a mixture of water glass and sodium hydroxide was applied as alkaline activator for the preparation of alkali activated aluminosilicates (AAA). A comprehensive analysis of the designed AAA included both functional and environmental aspects.

The best performance from a functional point of view exhibited the AAA material with the second lowest amount of alkaline activator, AAP70. It achieved the highest compressive and bending strengths despite of its highest open porosity. The compressive strength of AAP70 after 28 days, R_c, was 41.9 MPa, i.e., within the range of R_c values common for Portland cement pastes (22 MPa–66 MPa according to Su et al. (2015)). Therefore, the strength values could be considered competitive with cement based materials. The explanation of contradictory results obtained for the mechanical properties and porosity was found in the fine and homogeneous pore structure of AAP70, which was manifested in the highest amount of pores smaller than 1 μm and the lowest amount of pores larger than 100 μm.

The AAP70 showed the second lowest environmental impacts among the studied AAA, expressed in terms of consumed energy and emitted GHGs, which was due to the second lowest dosage of water glass and sodium hydroxide. In a comparison with the PCP having the compressive strength of 62 MPa (Pavlik et al., 2016), 63% of the total consumed energy and 81% of the emitted GHGs was saved which was very beneficial from the environmental point of view. The AAP60 paste with the best environmental parameters was not able to reach hardened state in a sufficient time and had to be excluded from further considerations.

The combined assessment of functional and environmental aspects, expressed using the energy consumption efficiency index and the CO_2 emission efficiency index per 1 MPa, showed that the best AAP70 mixture provided 45% savings in consumed energy and even 72% in the emitted GHGs, as compared with Portland cement paste.

Summarizing the results of the functional and environmental assessment of AAAs investigated in this paper, one can conclude that the utilization of waste brick powder generated by grinding of thermal insulation ceramic blocks in AAA is beneficial for three basic reasons: (i) its application in building industry presents an effective solution for disposal of a waste industrial product, (ii) its ready-to-use properties not requiring any thermal treatment or crushing further decrease its environmental impact, (iii) its use in AAA instead in cement-based materials leads to great reductions of consumed energy and emitted GHGs.

Acknowledgement

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3.3 Assessment of the reuse scenarios of waste materials

Selected journal paper

Fort, J., Cerny, R., 2020. Transition to circular economy in the construction industry: Environmental aspects of waste brick recycling scenarios. *Waste Management 118, 510-520.*

Viewing the recycling/reuse of waste materials as a viable scenario for the future sustainability of the construction industry, the comparison of particular scenarios represents a rule of the thumb to maximize the benefit. In this regard, four different scenarios for waste brick disposal (landfill, natural aggregate replacement, cement replacement, alkali activation) are accessed through Life Cycle Assessment analysis. As revealed, the most advantageous results in form of carbon dioxide emission reduction are attained by cement replacement and alkali-activation. On the other hand, the alkali activation significantly increased the damage to human health as a result of the application of alkaline activators.

Contribution to the practice: The presented conclusions point to the importance of the robust environmental analysis of building materials to avoid one-sided solutions and the transfer of burden from one area of protection to another.

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Transition to circular economy in the construction industry: Environmental aspects of waste brick recycling scenarios

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A B S T R A C T

The extensive exploitation of natural resources, together with an inefficient use of end-of-life materials, results in the generation of vast amounts of waste. The current material streams are to be reconsidered to mitigate the environmental burdens and achieve the sustainability goals. However, these intentions usually lead to material downcycling, which does not provide significant environmental benefits. In this paper, the potential of waste brick recycling is assessed from the environmental point of view as the recycling options of waste bricks attract an eminent attention due to rationalization and optimization of material streams, including transformation to the circular economy model according to the EU commitments. Three different scenarios are taken into account in that respect: replacement of natural aggregate, partial replacement of cement binder, and alkaline activation. The life cycle methodology is used at the assessment and the obtained results are presented on both midpoint and endpoint levels. The analysis of environmental impacts shows only minor improvements resulting from the replacement of natural aggregates by recycled waste bricks. The partial replacement of cement by waste bricks in powdered form can provide the most substantial benefits including decarbonization of the construction sector. The application of alkaline activators can harm the potential of alkali-activated materials considerably due to their negative effects on human health. A complex assessment of recycling scenarios is found to preferable to one-sided analyses aimed at carbon dioxide emission reduction only if a real sustainability without any hidden risks is to be achieved.

1. Circular economy in the construction industry

Since the building sector was recognized as a significant contributor to global carbon dioxide emissions, major efforts have been aimed at the mitigation of this effect (Açıkkalp et al., 2018; Bernal et al., 2018; Guardigli et al., 2018). A substantial portion of the consumed energy in the building sector is needed for material production, raw material gathering, high-temperature treatment, transportation, and waste disposal (Huang et al., 2018). The construction industry consumes enormous loads of virgin materials and the cement industry itself is responsible for the production of 8% of world carbon dioxide emissions (Miller et al., 2018). The increase of the world population goes hand in hand with increased demands on the production of new materials. The current linear model of the economy results in immense consumption of natural resources and dumping of pollutants at the end of the materials life cycle (Merli et al., 2018; Stephan and Athanassiadis, 2018). These factors present the main driving forces for the endeavor to deal with the negative consequences on the world climate.

The transition to a more efficient circular model of economics has ambitions to solve the sustainability problems on a higher level thanks to improved recycling and the creation of material loops (Horckmans et al., 2019; Zhang et al., 2019). Unfortunately, the change of this paradigm is accepted only by less than 10% of the world economy. Significant concerns with its wider acceptance are associated with the mismatch between national policies or poor political leadership (Rentschler and Bazilian, 2017; Williams et al., 2017). Another barrier can be seen in the limited practice and the not confirmed viability on a larger scale (Amaral et al., 2018; Kavlak et al., 2018). An example can be found in the recycling of construction waste (composed mainly of concrete and bricks), where the production requires significant energy inputs, but waste recycling is based on material crushing and the consequent utilization as aggregate, e.g., for road applications (Rodríguez-Robles et al., 2014). Therefore, the recycling of this energy-intensive material decreases the natural aggregate needs but, on the other hand, provides only limited environmental bene-

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fits, as compared with the original material fabrication (Pradhan et al., 2019, Horckmans et al., 2019).

The downcycling material strategy cannot be accepted as a progressive solution for the mitigation of negative environmental loads and transition to a circular economy. Dyllick and Rost (2017) presented significant advances from the point of view of products' or materials' sustainability concepts. They defined three sustainability models including both simple and advanced recycling concepts using a multilevel value (social, ecologic, economic). This concept provides an advanced assessment of sustainable production and raises the ambitions for both companies and society (Haarstad et al., 2018; Qian and Hamdany, 2015). Considering the sustainability principles and the present state-of-the-art, the exact characterization of environmental benefits supported by relevant functional characteristics of building materials needs to be established as a knowledge platform and decision supporting tool for stakeholders (Das et al., 2019; Yu et al., 2020).

The utilization of secondary or waste products is considered as a threshold parameter for the successful transition towards the circular economy model (Merli et al., 2018; Stephan and Athanassiadis, 2018). Despite the increased processing costs and energy consumption, the substitution of traditional products made from primary resources can deliver substantial benefits in the form of avoided production (Teh et al., 2017; Vitale et al., 2017). In other words, the total benefit is given by lower cumulative negative impacts. An additional benefit of the circular economy model presents the preservation of natural resources thanks to the landfilling restrictions and the centralization of industrial production (Fenner et al., 2018). The rethinking of the current paradigm is already apparent in several countries, such as Germany, Netherlands, Belgium, or Denmark, where up to 95% of the generated waste is reused or recycled (Ruiz et al., 2020). Notwithstanding, such an approach still faces downcycling even in the case of waste with high embodied energy. In a summary, the poor environmental viability of the downcycling prevents the achievement of a balance between all sustainability pillars, and leads to a slight improvement only(Fuhr et al., 2018; Reficco et al., 2018).

2. Recycling of waste bricks in current research: Functional and environmental considerations

While recycling of concrete is a frequently studied topic, recycling of brick waste was aimed predominantly at its use as a natural aggregate alternative (Ram et al., 2020, Pacheco-Torgal et al. 2020). The other options were either neglected or analyzed in a limited extent (Wong et al., 2018). Drawbacks associated with a low-value recovery of CDW can though impair the overall environmental benefits. Considering the available literature, three alternative utilization scenarios can be identified for waste bricks: replacement of natural aggregates, partial cement replacement, and total cement replacement using alkaline activation.

2.1. Replacement of natural aggregates

This option is the most often used one in the European region, especially due to its simplicity and low requirements for additional material processing and treatment. The processed waste brick (WB) is frequently used at road construction, which requires huge amounts of natural aggregates with not very demanding specifications. The recycled WB can be used as fine or coarse aggregate.

Many studies were focused on the preparation of recycled concrete using WB aggregate (Vieira and Pereira, 2015). Considering the mechanical performance of such concrete, a slight decrease in compressive strength and elastic modulus could be observed (Cachim, 2009) which - though was still acceptable for various construction applications. A more pronounced difference was found in the hygric properties of such recycled concrete; moisture conductivity and chloride ion penetration increased due to the high porosity of the brick aggregate (Dang et al., 2018). WB aggregates were also successfully used for the preparation of lightweight concrete with a bulk density below 1800 kg/m^3 (Zhao et al., 2018). The advantages of such material (excluding a lower environmental burden) consisted in improved thermal and acoustic insulation. Fine aggregates obtained by WB crushing were utilized for the development of new “green” mortars, plasters or unfired bricks (Dang et al., 2018; Hossain et al., 2019; Seco et al., 2018) but higher volumetric expansion was often observed when compared to control mixture. On the other hand, excessive moisture stored in the porous structure of brick aggregate reduced drying shrinkage (Wong et al., 2018; Zhang et al., 2018). Another positive effect of brick aggregate incorporation was described by Bektas et al. (2007), who reported that finely ground bricks reduced alkali-silica reaction in concrete mixtures and prevented durability loss.

The functional properties of cement composites containing recycled aggregates (RA) were studied frequently and were relatively well described in many research studies. Their environmental benefits consisting in the partial replacement of natural aggregates (NA) were analyzed less often in an explicit way. In one of such studies published recently Yazdanbakhsh et al. (2018) reported that the replacement of NA by RA did not have any substantial positive effect from the environmental point of view. Similar results were achieved also in some other studies, that revealed high sensitivity of overall results to transportation distances. A positive effect could be observed only when transportation distances were rather small and RA was used in higher volumes (above 20%). Downcycling of construction and demolition waste (CDW) or bricks to RA used mostly for road applications was characterized by Di Maria et al. (2018a) as a scenario with very low environmental benefits which came mainly from the avoided production of NA. The authors noted that the limited market for high-quality RA prevented more advanced recycling options and applications of such materials.

2.2. Partial cement replacement

The convenient chemical composition of waste bricks having a high content of SiO_2, Al_2O_3 and somewhat lower content of CaO makes good prerequisites for their application as supplementary cementitious materials if their grain size distribution is sufficiently fine (Afshinnia and Poursaee, 2015).

The pozzolanic reaction of brick powder was found mostly less intensive, as compared to the traditional supplementary cementitious materials, such as metakaolin or ground granulated blast furnace slag; it was due to the higher content of crystalline phase. However, sufficient grinding could increase the specific surface area and consequently also the pozzolanic activity (Čáchová et al., 2014; Komnitsas et al., 2015; Vejmelková et al., 2012). According to the subsequent experimental investigation of cement replacement in concrete, up to 20 wt% of cement could be successfully replaced without significant worsening of material performance (Vejmelková et al., 2012). The phase analysis revealed a dominant share (depending on the amount of used brick powder) of calcium silicate hydrates, which are commonly present in ordinary Portland cement mixtures, but calcium aluminate hydrates and calcium aluminosilicate hydrates were also found (Navrátilová and Rovnaníková, 2016). Aluminates and aluminosilicates in hydrated form can be considered as products of pozzolanic reactions responsible for the growth of mechanical strength at later ages but, on the other hand, an increased brick powder content can decrease the initial compressive strength and prolong setting times (Ge et al., 2012).

Partial replacement of cement in lime-cement mortars by brick powder was found as an effective solution leading to a substantial increase of compressive and flexural strength (Kočí et al., 2016). An overall positive effect of brick powder on the natural environment in the form of carbon emission savings was mentioned as well but no coherent analysis to support this fact was given.

Based on the available information, the utilization of ground waste bricks can be considered as a feasible scenario for bricks recycling and mitigation of negative externalities associated with bricks' end-of-life treatment but a detailed environmental assessment of this recycling option was not reported yet in common literature sources.

2.3. Total cement replacement using alkaline activation

Brick waste in powdered form can be used as a precursor for alkaline activation using sodium hydroxide or sodium silicate. A major advantage of this approach consists in the possibility to replace cement entirely, making the use of brick powder more effective than in the case of only partial cement replacement. The alkaline activation of brick or ceramic powder was studied less frequently in a comparison with slag or fly ash, which was due to its lower amorphous content. Prospective results were reported by several research groups only in the last decade (e.g., Amin et al., 2017; Fořt et al., 2018; Robayo-Salazar et al., 2017a).

Advanced mix design taking into account the importance of various molar ratios (particularly SiO_2/Al_2O_3, Al_2O_3/Na_2O, and SiO_2/Na_2O) confirmed the suitability of brick powders to replace traditional binders and preserve the mechanical performance of designed materials. Besides the environmental benefits, alkali-activated materials showed excellent chemical and high-temperature resistance, which supported their use in specific applications. A substantial knowledge base in this field was laid by Reig et al. (2013a), who showed the usability of red brick waste for alkaline activation and reached a compressive strength of ~ 30 MPa. A further improvement of mechanical parameters was done in the consequent research by optimization of SiO_2/Na_2O ratio when 50 MPa was achieved (Reig et al., 2013b). Robayo-Salazar et al. (2017b) investigated the effect of elevated temperature curing on residual properties of alkali-activated red bricks with Portland cement addition and reached compressive strength up to 102 MPa when 20 wt% of cement was applied. Considering the environmental effectiveness in terms of carbon dioxide production, the best results were obtained for mixture with 10% of Portland cement having compressive strength of ~ 48 MPa and carbon dioxide production reduced to about one half as compared to traditional concrete.

Fořt et al. (2018) analyzed both functional and environmental aspects of alkaline activation of brick powder using energy efficiency and CO_2 emission efficiency indexes and found a substantial positive effect of waste bricks valorization. The environmental benefits related to the replacement of traditional binders by alkali-activated materials were discussed also by McLellan et al. (2011), who underlined the importance of transportation for the overall results. The contribution of alkaline activation to carbon mitigation strategies was questioned by Habert et al. (2011), but considerable improvements can be achieved were achieved when one part geopolymers were used (Ouellet-Plamondon and Habert, 2014)

2.4. Contribution of this study to the state of the art

Although the utilization of waste materials in materials engineering has attracted a significant attention over the last decades, the recent knowledge related to waste bricks recycling remains highly fragmented. In particular, there is a need for a comprehensive environmental assessment, which can provide a coherent outcome with clearly highlighted focal points utilizable for future research and policy implications. This study is focused on a detailed and complex comparison of the environmental impacts of various waste brick recycling scenarios based on the life cycle assessment to avoid one-sided conclusions. Such research was not reported yet in common literature sources.

3. Eco-efficiency of waste brick recycling: Applied methodology

3.1. Goal of the study and functional unit

This study evaluates the environmental impact of the scenarios of waste brick disposal or recycling characteristic for the European area. For the comparison of studied scenarios, the life cycle assessment methods (LCA) are employed. The functional unit used in this study is 1 metric ton of waste red bricks, discarded bricks, or waste formed during brick production (Fořt et al., 2019, 2020), which are currently considered as waste materials with limited use. For better clarity and clear comparison with other scenarios, functional parameters were taken into account as an implicit condition for intended mixture design based on the achieved results of (Kočí et al., 2016; Vejmelkova et al., 2018).

3.2. Scenarios and system boundaries

Three alternative recycling scenarios for the waste brick recycling were considered, namely the utilization of WB for recycled aggregate production (RA scenario), partial cement replacement in composite materials (CR scenario), and alkaline activation for precast blocks production (AA scenario). In order to complete the list of scenarios, the landfilling option (LA scenario) was also taken Into account despite the fact that this alternative is not applicable in a number of countries due to the legislative restrictions. The analysis takes into account regional factors of the Czech Republic, but with slight modifications it can be applied for other central European countries. The Czech Republic, compared to substantially larger countries such as Italy, France or Germany can use shorter transportation distances. Considering the national energy mix, a relatively high share of electricity is produced by coal and natural gas (57%), followed by 39% from nuclear plants and only about 4% from renewable energy plants. A detailed information on each scenario is given below.

3.2.1. Landfilling (LA)

This scenario is considered for a comparison only, due to the abandoned landfilling in several countries (Das et al., 2019; Horckmans et al., 2019; Silvestre et al., 2014). In the case of the LA scenario, the entire redundant material including the brick powder and stony fraction is collected, stored near the production plant, and then taken away to dump.

3.2.2. Recycled aggregate (RA)

The utilization of waste bricks in the form of recycled aggregate can be considered as the most often used scenario for the countries where landfilling is abandoned or highly-priced in particular. While the larger fraction can be used for concrete production, the application of fine fraction as road subbase prevails (Rodríguez-Robles et al., 2014; Yazdanbakhsh and Lagouin, 2019).

The RA scenario consists of several successive steps including collection, transport, separation, crushing, and milling to obtain more homogenous ready-to-use material without impurities (see Fig. 1). The recycled aggregate scenario considers transportation of CDW from the demolition site to the treatment plant, where consequent processing of debris takes place. The devices used for

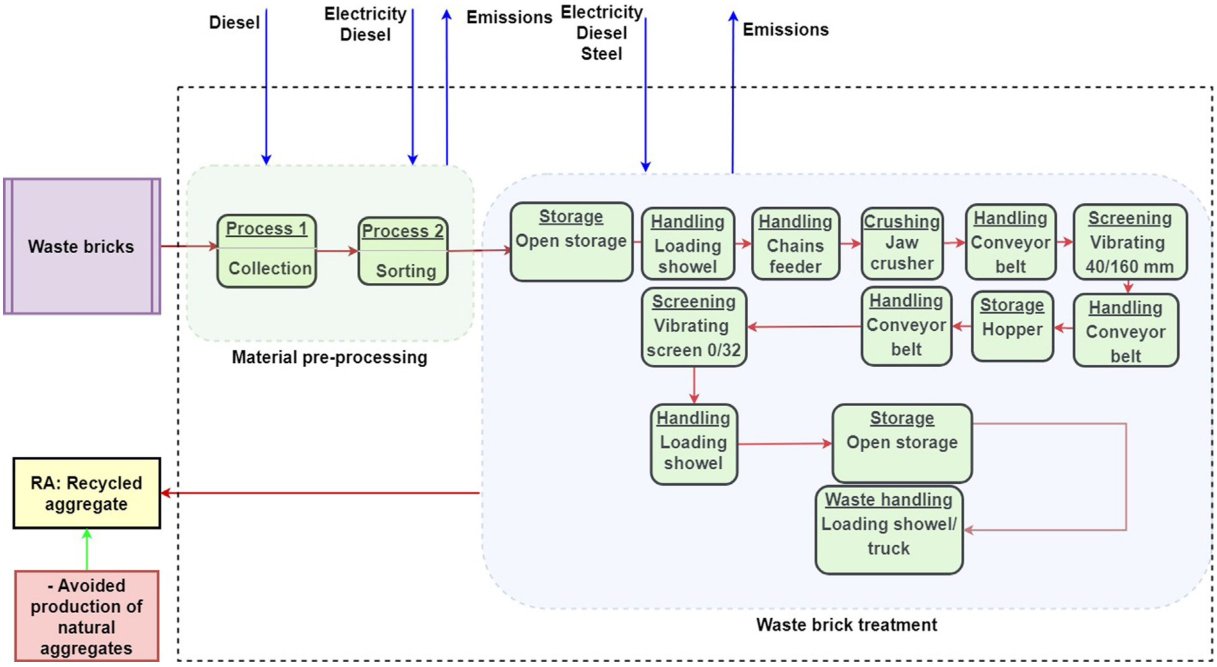

Fig. 1. Flowchart of recycled aggregate (RA) scenario.

the RA production are similar to the machinery employed for NA but additional processes, such as separation of plastics, ferrous metals, and other waste, are included (Ghanbari et al., 2018; Rosado et al., 2019). Typically, loading shovels, chain feeders, jaw crushers, conveyor belts, and vibrating devices powered by diesel or electricity are used to produce recycled aggregate. The selective demolition is not taken into account in the present study despite its substantial beneficial effect on the impurities volume (Di Maria et al, 2018b; Cuenca-Moyano et al., 2019).

Natural aggregates are substituted in this case and their production is avoided. The recovery ratio for recycled aggregate production is about 98% for fine aggregates.

3.2.3. Partial cement replacement (CR)

In this scenario, the material processing presents an extension of the process described in Section 3.2.2 because a finer fraction, typically with d_{50} of ~50 μm, must be produced. For this purpose, additional milling and vibration are employed (see Fig. 2). The last important step aimed at the achievement of pozzolanic activity of such material comprises drying at 70 °C to remove excess water (Pavlík et al., 2016; Calvo and Domingo, 2017). The obtained material can be utilized as a replacement of energy-intensive traditional binders in plasters or concrete (Kočí et al., 2016; Vejmelkova et al., 2018).

3.2.4. Alkaline activation (AA)

Similarly to the partial cement replacement scenario described in Section 3.2.3, the waste brick needs to be processed at first to obtain a fine fraction with $d_{50} \sim 50$ μm and then dried. Additionally, alkaline activators (e.g., sodium hydroxide, sodium silicate) are used in the mix design (Fořt et al., 2018). The energy consumption for material mixing and molding is also taken into account. The mixture analyzed in this study contains sodium hydroxide pellets, sodium silicate (water glass) with SiO_2/Na_2O molar ratio of 1.6, sand, and finely milled brick dust (see Fig. 3) and provides mechanical performance comparable with common cement-based composites (Fořt et al., 2018; Robayo-Salazar et al., 2017b).

3.3. Life cycle inventory

The life cycle inventory includes all inputs and outputs for the particular scenarios described above. All related data sources are shown in the Supplementary Material, Table S1 with the relevant references. The obtained data were modified according to the functional unit and included in the Simapro LCA software 8.5. The data for raw material production, processing, transport emission, and energy production were extracted from the Ecoinvent database v.3.5. The Czech energy grid with a dominant share of coal energy sources was considered. The consumption of auxiliary materials, such as water for the dust control or steel for machinery maintenance and lubricants, was taken into account. Since the discarded bricks present waste material, no environmental footprint associated with waste brick production was assumed; only transportation, collection, and consequent processing impacts were considered. For the preparation of alkali-activated mixtures, no data in Ecoinvent was available. Therefore, the data published in the scientific literature was used.

3.4. Avoided production

In all alternative scenarios, landfilling is prevented, which can be considered as a benefit attributed to recycling. However, it cannot be included in LCA as avoided production according to the European standards. The reasons and rules associated with the calculation of alternative use of end-of-life building materials are analyzed, e.g., in Silvestre et al. (2014). Therefore, only the material aspects of avoided production, such as replacement of natural aggregate or cement in various concrete products (precast concrete blocks, concrete mixtures), are taken into account.

The substitution level of original materials for the particular scenarios was derived from the study of Borghi et al. (2018). This method is based on the assessment of recycled material quality using its composition (Q1) and functional properties (Q2). To provide a more appropriate substitution ratio, the market perception ratio (M) was included in the calculation. The substitution coefficient (S) was then defined as

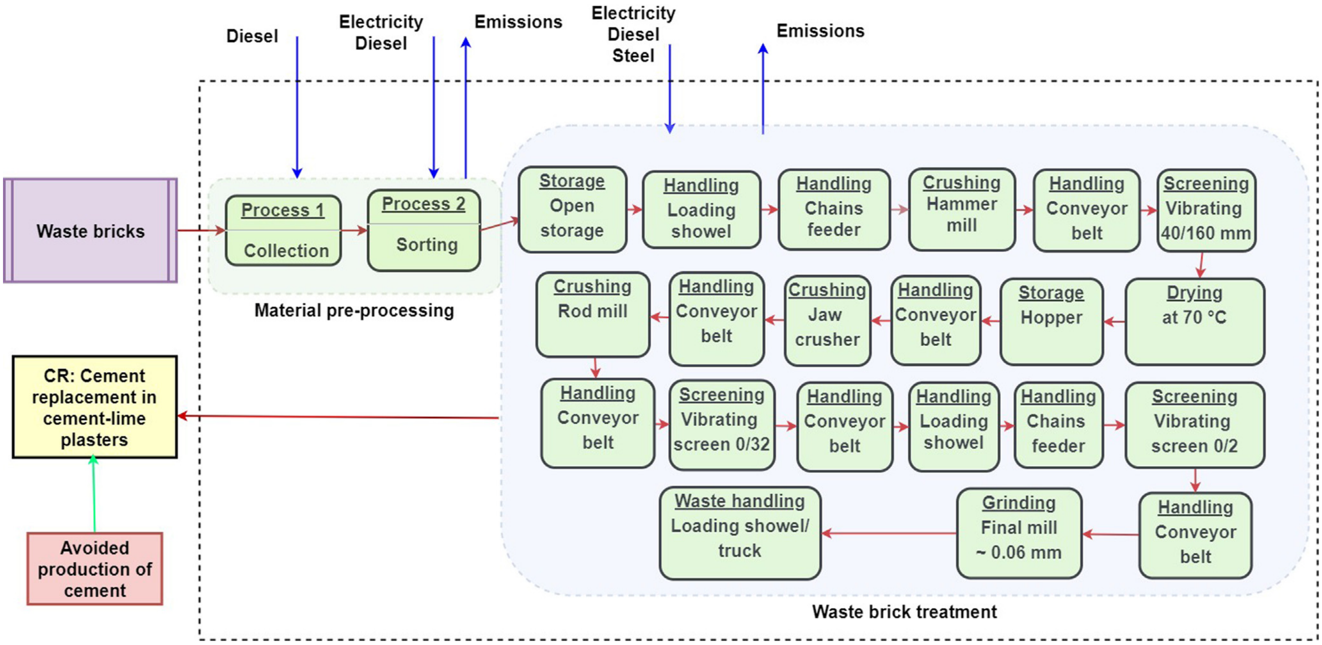

Fig. 2. Flowchart of cement replacement (CR) scenario.

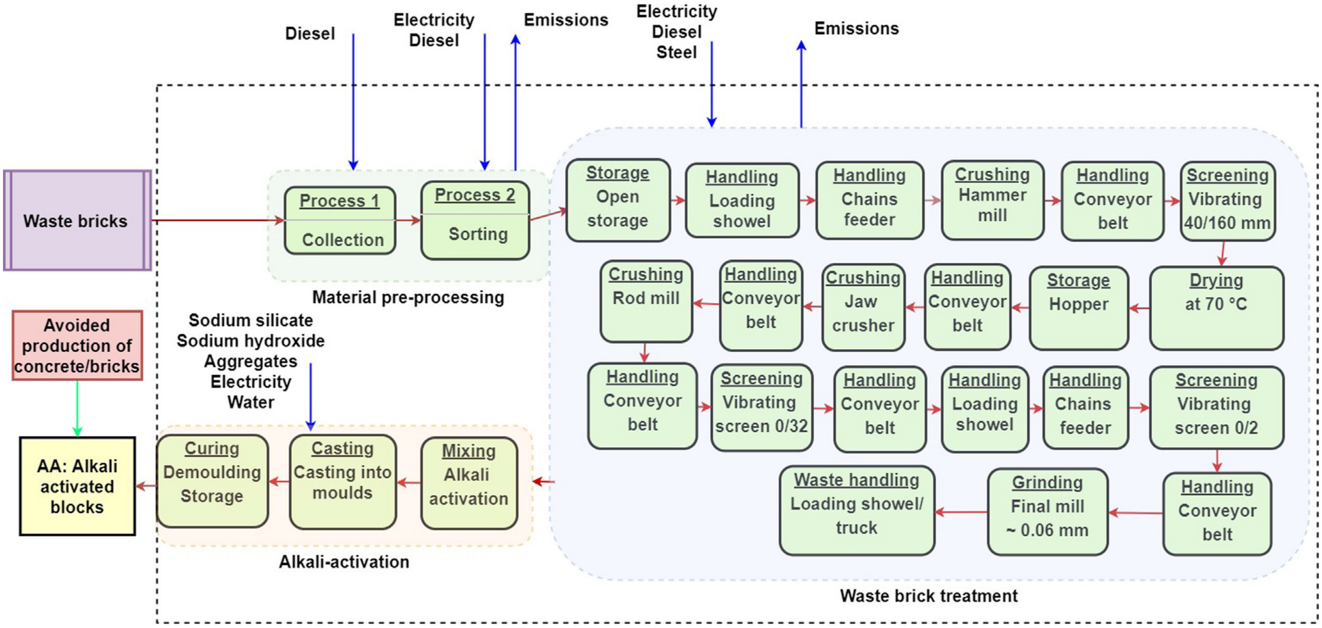

Fig. 3. Flowchart of alkaline activation (AA) scenario.

$$S = Q_1 \cdot Q_2 \cdot M$$

Whilst the quality of a material can be easily assessed using appropriate experimental procedures and analytical devices, the estimation of market perception is more difficult as its indicators are not clearly defined. The fundamental barriers for a more positive acceptance of alternatives for a traditional product are usually based on the lack of knowledge, different application procedures, and distrust. On the other hand, decreased prices and sufficient availability can substantially improve the market acceptance of alternative products.

In this study, the substitution coefficients were calculated on the basis of quality parameters and market demands found in the available literature sources (Azúa et al., 2019; Bassani et al., 2019; Borghi et al., 2018; Meng et al., 2018; Wong et al., 2018). The S values obtained for the RA, CR and AA scenarios were 0.83, 0.71 and 0.65, respectively.

3.5. Transportation distances

Transportation causes substantial environmental loads particularly in the case of aggregate or other low-performance materials production (Di Maria et al., 2018a, 2018b; Ghanbari et al., 2018; Yazdanbakhsh et al., 2018). Considering the locations of current waste bricks landfills and production plants, relatively short distances for material transportation ranging from 20 to 50 km were assumed in this study. As most of the produced waste is transported by lorries having loading capacity up to 23 t, this transportation option was used in the analyses.

3.6. Life cycle impact assessment

In this study, the Impact 2002 + methodology (version 3.5) by Simapro 8.5 was used for the calculation of midpoint and endpoint indicators. This methodology was chosen mainly because of its suitability for the European region. In addition, the complex list of 15 impact categories provided a robust platform for the identification of environmental burden on both midpoint and endpoint level (Owsianiak et al., 2014).

The impact categories included in the assessment were as follows: Aquatic acidification (AAC), Aquatic ecotoxicity (AE), Aquatic eutrophication (AEU), Carcinogens (CA), Global warming (GW), Ionizing radiation (IA), Land occupation (LO), Mineral extraction (ME), Non-carcinogens (NCA), Non-renewable energy (NRE), Ozone layer depletion (OLD), Photochemical oxidation (PO), Respiratory effects (RE), Terrestrial acidification/nitrification (TAN), and Terrestrial ecotoxicity (TE). For the comparison of different scenarios, the environmental midpoint impact categories were merged to estimate the endpoint impact categories, which included human health, ecosystem quality, climate change, and resources. The obtained results were normalized into a single-score form to achieve a clear and explicit comparison.

3.7. Sensitivity analysis

In order to provide a robust and comprehensive model of environmental impacts of waste bricks recycling, the sensitivity analysis at the endpoint was carried out, in terms of uncertainty, for the most critical inputs. This analysis contributes to the understanding of how the different values of input parameters influence the system. The substitution ratio for the avoided production in each scenario and transport distances were the main parameters considered in the analysis. For the calculation of outputs' sensitivity to the avoided production, the variation steps of 5% for substitution ratio and 10% for transport distances were used. The detailed parameters for the particular options are summarized in Supplementary Material Table S2.

4. Results

4.1. Midpoint level

4.1.1. Comparative assessment

Fig. 4 shows the results obtained on the midpoint level. The environmental indicators are presented for all categories as impacts relative to 100%. Apparently, the most significant benefits were achieved for carcinogens (CA), ionizing radiations (IR), respiratory organics (REO), and global warming (GW) in the AA scenario. Distinct benefits could be found for non-carcinogens (NCA), mineral extraction (ME), and global warming (GW) in the CR scenario, too.. However, it should be pointed out that despite the positive effect on the global warming manifested by the low carbon dioxide equivalent production (Abdallah et al., 2016)this category should not be preferred at the expense of others. A too narrow assessment of industrial production may result in possible negative consequences in the future. For AA it was well documented by the results obtained for mineral extraction (ME), aquatic ecotoxicity (AE), aquatic eutrophication (AU), land occupation (LO), and respiratory inorganics (REI) that were substantially worse than for the other scenarios. Discrepancies between the particular impact categories were revealed also for CR and RA scenarios. Negative impacts were observed for non-carcinogens (NCA), non-renewable energy (NRE) and terrestrial ecotoxicity (TE) in the RA scenario, for ozone layer depletion (OLD) and respiratory organics (REO) in the CR scenario. Landfill abandoned in several countries exhibited the worst impact on aquatic ecotoxicity.

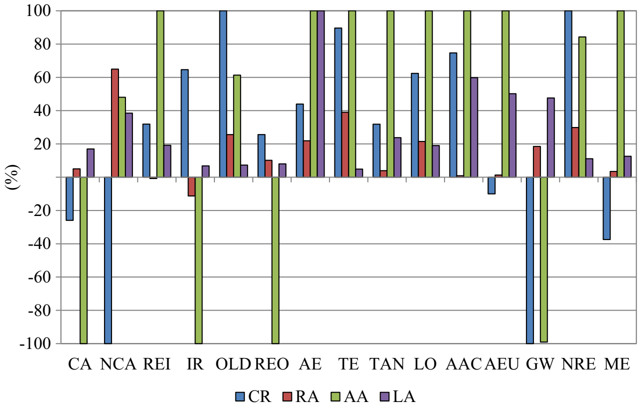

Fig. 4. Comparison of particular waste brick scenarios.

Considering the benefits of avoided production, the RA scenario did not result in any distinct advantages related to avoided natural aggregate production, since the brick treatment, sorting, crushing and transport exceeded the gained improvements. It should be noted that the viability of using RA of various origin was found limited also by other investigators (Ghanbari et al., 2018; Pradhan et al., 2019; Wong et al., 2018). The main reason was that natural aggregates production does not require energy-intensive treatment processes. On the other hand, noticeable environmental savings were achieved by avoided cement or concrete production in the case of CR and AA scenarios which was due to high energy consumption associated with the material production using original resources. These findings are in a qualitative agreement with Di Maria et al. (2018a) who characterized the high-quality recycling of CDW as a powerful tool for significant reduction of negative environmental impacts associated with building materials production.

4.1.2. Contribution analysis

(a) RA scenario

The midpoint environmental impacts relative to 100% calculated for the RA scenario are given in Fig. 5. The most distinct benefits of avoided production were identified for ionizing radiation (IA), respiratory inorganics (REI), and mineral extraction (ME) whilst the other gains ranged up to 30%. Nevertheless, the negative impacts prevailed, as compared with the positive impacts related to avoided production. A major contribution to the considered impact categories presented material crushing and grinding processes employed to obtain the desired aggregate fraction. Therefore, substantial energy savings could be reached when waste brick powder produced at bricks crushing and cutting was utilized in part (Fořt et al., 2018). However, the inclusion of another transportation may result in worsening some impact categories, which are highly dependent on transportation distances, ionizing radiation (IA) and respiratory organics (REO) in particular (Di Maria et al., 2018a). The remaining treatment processes, such as sieving, sorting, and loading, exhibited only minor impacts on the obtained results and did not represent any significant barrier. In a comparison with the results published by other investigators, the environmental impacts calculated in this study partially agreed with Borghi et al. (2018), who reported savings of natural resources but an increase of carbon dioxide emissions due to the low-grade

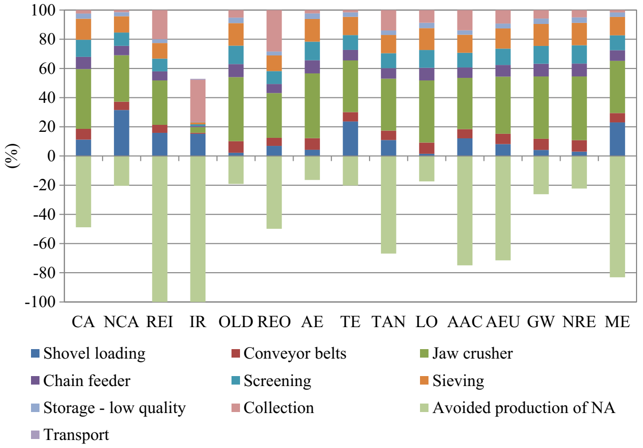

Fig. 5. Midpoint impacts for RA scenario.

applications. Yazdanbakhsh and Lagouin (2019) admitted the environmental benefits related to recycled coarse aggregate production when the landfill option was considered as an avoided product.

(b) CR scenario

The quantified potential environmental impact related to the replacement of cement in plasters by waste bricks in powdered form is presented in Fig. 6. Compared to the RA scenario, even higher environmental profit from the avoided was achieved in global warming (GW), mineral extraction, carcinogens, non-carcinogens, and aquatic ecotoxicity. On the other hand, respiratory inorganics (REI) and ionizing radiation (IR) revealed worsened values. A substantial impact on the increased environmental burden was associated with bricks grinding and drying in particular.

It should be pointed out that the overall environmental benefits of this scenario include also a positive balance of CO_{2eq} when considering the carbon–neutral economy targets (Allwood et al., 2010). Contrary to some other studies aimed at the replacement of cement in concrete mixtures, the analyzed replacement of cement in plasters does not struggle with functionality restrictions (Abed and Nemes, 2019; Lin et al., 2010; Mo et al., 2017) and provides a competitive alternative to cement-lime plasters. An additional benefit can be found in the application of such plasters for the restoration of historical buildings where cement-lime plasters are not in line with the historical context (Koci et al., 2010; Liu et al., 2017). The replacement of cement in low-performance concrete should be considered in future studies as a prospective option for the waste bricks recycling (Naceri and Hamina, 2009).

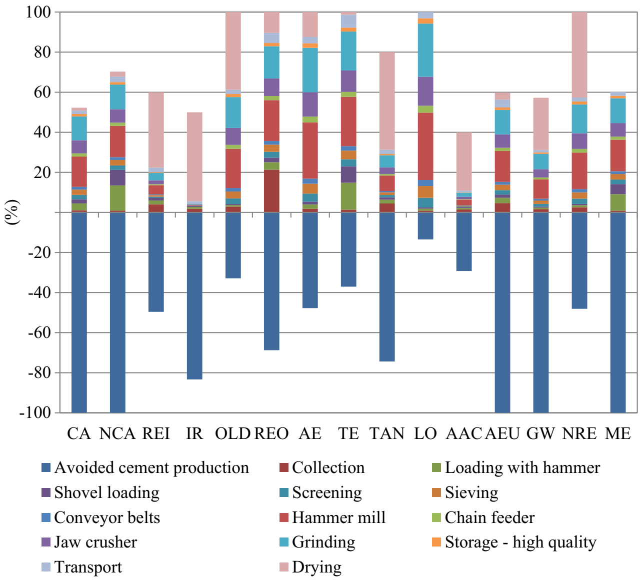

Fig. 6. Midpoint impacts for CR scenario.

(c) AA scenario

The process and materials contribution analysis of the utilization of waste brick for the production of AA precast blocks and AA mixtures are given in Fig. 7. The sum of avoided impacts was noticeable for global warming (GW), ionizing radiation (IA), carcinogens (CA), and respiratory organics (REO) in particular. The production of used alkali activators substantially impaired the benefits related to the avoided production of ordinary concrete blocks or fresh concrete mixtures. Compared with sodium silicate, the environmental burden associated with sodium hydroxide did not reach the same level, which was due to the considerably lower sodium hydroxide dosage in the AA materials design. Notwithstanding, Samarakoon et al. (2020) and Vinai and Soutsos (2019) already reported on alternative ways to reduce dosages of energy-intensive activators. Besides the environmental loads of applied activators, grinding and drying had a substantial negative impact, while the rest of the processes resulted in only minor environmental damage. Remaining categories exhibited greater negative connotations despite the relatively low transport distances, which are often viewed as a barrier for alkali-activated materials viability (McLellan et al., 2011). From the point of view of carbon dioxide production, which is the most pursued category, significant savings can be reached by using AA materials, as it was concluded by several authors (Mehta and Siddique, 2018; Provis, 2018; Robayo-Salazar et al., 2017b; Salas et al., 2018; Talang and Sirivithayapakorn, 2018). However, these gains may be accompanied by the worsening of other impact categories, as it was described in the analysis presented in this paper.

4.2. Endpoint level

4.2.1. Comparison on the endpoint level

The normalized endpoint impacts of all analyzed scenarios are presented in Table 1. Similarly, to the midpoint level, different environmental impacts were obtained for particular categories.

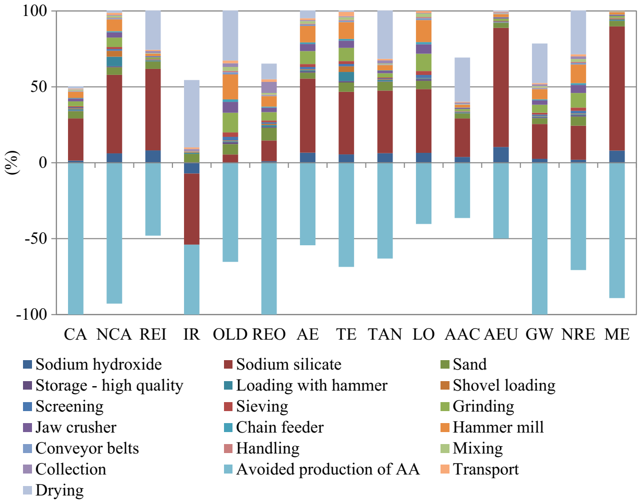

Fig. 7. Midpoint impacts for AA scenario.

The CR and AA scenarios showed very prospective results for the mitigation of climate change thanks to the negative balance in the production of carbon dioxide equivalent.. On the other hand, the AA scenario extensively increased the environmental burden on human life. For the ecosystem quality, all scenarios had a similar impact with only minor variations. The highest negative impact associated with resource consumption was found for the CR and AA scenarios since the advanced processing of waste bricks required additional assets. The RA scenario was found as the least demanding alternative if ecosystem quality was taken into account.

4.2.2. Normalized endpoint assessment

The weighted and normalized endpoint damage assessment is shown in Fig. 8. The RA scenario resulted in the least impact on all endpoint categories but no environmental benefits were identified. It was not found favorable even when compared to landfilling. Notwithstanding, the material landfill is not applicable in several European countries and cannot be considered as avoided production (Silvestre et al., 2014). Significant benefits were achieved for the CR and AA scenarios in the area of the impact on climate change represented by the CO_2 equivalent. Both scenarios provided a negative balance of CO_2 production thanks to the avoided production of energy-demanding products, such as concrete blocks or Portland cement. On theother hand, substantial drawbacks related to the impact on human health were observed, particularly in the case of the AA scenario. Both scenarios were also demanding on resources consumption as a result of more advanced processing.

4.3. Sensitivity analysis

The results of sensitivity analysis for particular scenarios, which was performed using the parameters given in Supplementary Material Table S2, and taking into account processes with the highest uncertainty, are presented in Supplementary Material Fig. S1 - 3 in the form of the endpoint single score given in milipoints (mPt).

The RA scenario (Supplementary Material Figure S1) showed only a minor effect of the substitution ratio on the overall environmental impact, contrary to the other two scenarios that could be attributed to the lower quality requirements on the aggregates.. On the other hand, the effect of different transport distances exhibited a more significant sensitivity, which complied with the findings of Di Maria et al. (2018a) and Yazdanbakhsh et al. (2018). As a result, the overall environmental score varied from 6.3 mPt to 13.56 mPt.

The consequences of variations in the substitution ratio were important for the CR and AA scenarios in particular. The sensitivity analysis of the CR scenario (Supplementary Material Figure S2) showed a substantial variation when the substitution ratio was modified. Here, the environmental score varied from 8.1 to 18.2 mPt, which, compared to the alteration of transport distances, represented a substantially more sensitive input parameter. The most variable results were obtained for the AA scenario (Supplementary Material Figure S3) where even small changes in substitution ratio and sodium silicate dosage resulted in substantial worsening or improvement of the overall environmental score. The sensitivity to the alteration of transport distances was for both CR and AA scenarios insignificant.

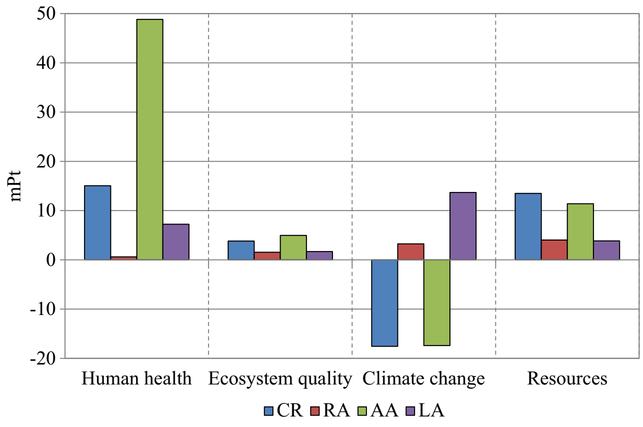

Fig. 8. Comparison of different scenarios on endpoint level.

5. Discussion

The analysis of the environmental impact of three different waste brick recycling scenarios presented in this paper confirmed that waste material recycling poses a challenge for the current research. In particular, the alteration of current material streams should be reconsidered to mitigate the consequent environmental burden. The inefficient use of materials resulting in massive depletion of natural resources and generation of vast amounts of waste materials can be solved by adopting the circular economy model that prefers recycling to landfilling (Das et al., 2019; Huang et al., 2018).

However, such intentions usually led to downcycling only to date, which did not provide significant improvements due to the limited environmental benefits. A possible further step in promoting more advanced recycling options may be seen in the legislative support of the production of recycled materials having satisfactory functional properties, comparable with the original ones, which can improve the market acceptance ratio (Ghaffar et al., 2020; Hasan et al., 2020; Nylén and Salminen, 2019; van Deventer et al., 2012).

Taking into account the outdated European building stock (Asdrubali et al., 2019), energy-demanding thermal comfort maintenance of residential buildings, and the actions towards the refurbishing of old houses to meet the criteria of nearly-zero energy buildings (NZEB), there are huge needs for new building materials. In this sense, the current building stock should be considered as a material bank that can be effectively utilized in the form of valorization of “waste materials” into new advanced materials that can completely replace the raw materials. This action requires a robust database of not only functional parameters, such as mechanical, hygric and thermal properties. The environmental studies need to be involved as well to provide the most sustainable solutions (Amato et al., 2019; Bernal et al., 2018).

Table 1
Endpoint damage assessment.

Damage category	Unit	CR	RA	AA	LA
Human health	DALY	1.07–04	1.45E-06	3.46E-04	3.87E-05
Ecosystem quality	$PDF^*m^{2*}yr$	52.2	21.2	67.9	38.77
Climate change	kg CO_2 eq	−174	32.2	−172	132.81
Resources	MJ primary	1364.54	612	1224.42	377.31

The downcycling of waste brick into the recycled aggregate provides a viable solution for waste brick recycling due to the low requirements on material processing. Further processing steps increase the energy demands but they also provide a substantially improved environmental performance related to the benefits of avoided production. Additionally, a comprehensive environmental assessment should be preferred to the use of single criteria, such as the carbon dioxide production, to avoid one-sided conclusions. In other words, the obtained results must be presented in a more complex way to prevent another risks since the desired recycling scenario must comply with all sustainability pillars.

In the last decades, alkali-activated materials have been extensively studied as a prospective solution having high ambitions to replace traditional concrete production (Asim et al., 2019; Luukkonen et al., 2018). In fact, carbon dioxide emissions were notably decreased but other negative consequences of industrial production were often not included in the LCA analyses which made these materials a controversial option. The benefits associated with alkali-activated materials were questioned in several studies where negative environmental consequences of the intensive NaOH, KOH, or Na_2SiO_3 use were found (Abdulkareem et al., 2019; Gavali et al., 2019). The real potential of alkaline activation of waste bricks remains though uncovered due to the prevailing application of energy-intensive alkaline activators, such as sodium hydroxide or sodium silicate. The sensitivity analysis of the AA scenario presented in this paper showed how even small variations in activators' dosage can influence the total environmental impact. Therefore, a finer adjustment of their dosage or possible replacement by other suitable alkaline substances presents a subject worthy of further studies from both technological and environmental points of view.

It should be noted that the results obtained in this study have some limitations which may though be considered as an inspiration for future work. Contrary to the RA scenario, the CR and AA scenarios are currently not practiced on the industrial level. A broader utilization of such scenarios requires the optimization of processes involved that can provide additional benefits (Evrard et al., 2016). Such factors are, however, not included in the analysis.

6. Conclusions

Facing the lack of original materials, the environmental and economic externalities associated with raw material quarrying, and the requirements paid on mitigation of produced waste, red brick waste has been extensively studied as alternative concrete-making material from a functional point of view (Wong et al. 2018). However, the current research papers do not provide a robust assessment of environmental impacts as one of the sustainability pillars required for a long-term viable solution.On this account, three potential waste brick recycling scenarios were compared, and their environmental impact was analyzed using the LCA methodology. The most important results can be summarized as follows:

The application of waste bricks as a recycled aggregate for concrete mixtures, subbase, or road pavement (RA scenario) did not provide noticeable environmental benefits to carbon dioxide reduction, despite its lowest environmental impact score. The main argument for this conclusion was in only minor savings associated with the avoided production of natural aggregates.

The utilization of waste bricks in powdered form as partial replacement of Portland cement (CR scenario) represented the most favorable option with substantial environmental benefits and limited negative consequences.

The alkaline activation of waste brick powder (AA scenario) was found controversial from the environmental point of view. It was very beneficial as for the carbon emissions but, it showed negative impacts on human health.

The valorization of brick waste can be considered as a viable solution only if a complex technological and environmental characterization is done.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2020.09.004.

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3.4 Lowering the environmental profile of alkali-activated materials

Selected journal paper

Fořt, J., Mildner, M., Keppert, M., Černý, R. 2022. Waste solidified alkalis as activators of aluminosilicate precursors: Functional and environmental evaluation. *Journal of Building Engineering, 54, 104598.*

The design of alkali-activated materials is accompanied by the utilization of the alkaline solution, usually based on sodium hydroxide, sodium silicate, or potassium hydroxide. However, the commercial production of these alkaline solutions is energy demanding and thus harms the natural environment despite the considerable carbon dioxide savings. To prevent the transfer of the burden to another area of protection, the replacement of the commercial alkaline activator, the waste alkalis disposed of in the glass industry was studied as an alternative to a commercial one. Obtained results refer to the viability of this approach in terms of meeting sustainable principles and reduction of the environmental burden. Within the material description, the combined integrated assessment is proposed and employed.

Contribution to the practice: *This study reveals the potential of locally available waste alkalis as a replacement for energy-intensive and expensive commercial alkaline activators as a way toward reduction of the environmental footprint of alkali-activated materials as well as the volume of produced hazardous waste.*

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Waste solidified alkalis as activators of aluminosilicate precursors: Functional and environmental evaluation

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ABSTRACT

Alkali-activated materials research presents currently a hot topic in materials science mainly because of the potentially significant savings of carbon dioxide emissions compared to Portland cement. A broad range of precursors was studied to highlight the environmental and functional benefits related to the utilization of local materials or waste products. However, the negatives associated with the application of commercial alkaline activators, such as sodium hydroxide or sodium silicate, which can pose a significant environmental risk in some impact categories, were often overlooked. In this study, the potential of waste solidified alkalis originated in industrial operations as primary activators of aluminosilicate precursors is analyzed. At the functional and environmental evaluation of designed alkali activated aluminosilicates, basic physical characteristics and mechanical parameters represent the functional properties, the LCA analysis at midpoint and endpoint levels describes the environmental impacts. The mechanical performance is found satisfactory for construction materials, the comparison of environmental parameters with widely used commercial activators shows substantial benefits boosted by the avoided production of neutralization agents used for the stabilization of waste alkalis.

1. Introduction

The rapid population boom accompanied with the urbanism development intensifies the pressure on “greening” of building materials since the Portland cement production is responsible for about 15% of total industrial energy and about 8% of total carbon dioxide emissions [1]. Facing the environmental consequences from extensive carbon dioxide production and natural resources exploitation, incentives for the use of alternative fuels at clinker production, replacement of concrete aggregates, and replacement of cement by supplementary cementitious materials have been introduced [2–4]. However, the fuel alteration does not provide sufficient improvements, since the major volume of emitted carbon dioxide arises from limestone calcination [5]. The replacement of natural aggregates by recycled materials originated within industrial activities represents a very popular effort aiming at concrete “greening”. In this regard, benefits related to the preservation of natural resources can be found. Notwithstanding, the overall environmental burden is reduced only slightly due to the high sensitivity of the environmental score on transportation distances [6]. Similar conclusions were formulated also by Refs. [7,8] who highlighted the importance of the local availability of used recycled aggregates. Among others, cement replacement attracted substantial attention from the scientific community within the last decades [9]. The utilization of pozzolanic reaction of various industrial by- and waste products contributes to the preservation of the mechanical

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parameters but it provides satisfactory results approximately up to 30% replacement only [10,11]. In other words, this strategy may provide only partial improvements, which, unfortunately, are not sufficient, given the requirements of long-term greenhouse emission development strategy of the EU [12].

Binders based on alkaline activation represent an effective approach due to the significantly increased amount of utilized waste materials, as well as the avoided Portland cement production [13]. Recently, the use of alkali-activated materials was found as an effective strategy heading to the development of sustainable building materials with sufficient mechanical parameters at reduced carbon dioxide emissions [14,15]. The environmental efficiency and eco-friendliness of alkali-activated materials strongly correlate with the type of the selected source material, as well as the dosage of used alkaline activators [16]. In this regard, various natural precursors, such as clay and kaolin, agricultural waste (rice husk ash, palm oil ash), and industrial waste or by-products (silica fume, blast furnace slag) were investigated [17–20]. As revealed, the binders realized through the alkaline activation of selected precursors by sodium hydroxide, sodium silicate, potassium hydroxide, or their combination exhibited sufficient compressive strength, increased durability, and fire resistance, and faster setting time compared to cement-based binders [21]. Since the environmental benefits assigned to alkali-activated binders arise from completely avoided Portland cement production and valorization of various industrial by- or waste products, the utilization of natural precursors cannot be deemed as a sustainable solution due to natural resources depletion with results in worsened environmental scores [22]. Considering the outcomes of performed studies, a reduction in carbon dioxide emission ranging from 30 to 65% can be expected [23,24]. On the other hand [16], questioned the environmental efficiency of these strategies due to substantial transportation distances that increase the environmental costs. Moreover, the life cycle assessment results are influenced by the used alkaline activator in terms of the type, dosage, production technology, etc. It should be noted that the production of both sodium silicate and sodium hydroxide is the energy-demanding process that results in a high environmental burden [25,26]. The relevance of the applied alkaline activator on the overall environmental score was mentioned in the work of [27], who concluded on a high sensitivity of environmental savings to the type and dosage of alkaline activator. While the major positives arise from carbon dioxide emission savings secured by utilization of large volumes of solid precursors, the negative impact of first grade alkaline activators use remains underestimated. Besides the high carbon dioxide release during their production, severe adverse effects notable in other impact categories (fresh water ecotoxicity, particulate matter, ionizing radiation, etc.) can be found [26,28].

To avoid one-sided solution, attempts to replace first-grade activators with environmentally friendly alternatives comprised the utilization of waste glass cullet [26,29], or biomass ash rich in potassium [30,31]. However, the questionable availability of waste glass diminished the viability of this approach. Taking into account the glass recycling rate of about 78% and target of 90% by 2030, the very limited availability of rice husk ash in European countries, new perspectives for waste-derived alkaline activators need to be introduced [32]. In these activities, local availability plays a crucial role, and national industrial activities can be viewed as possible sources of alkalis [33]. Nowadays, glass manufacturing, beverage production, and textile plants are the most significant industrial sources of waste alkalis. Such waste materials are labeled as hazardous waste that requires special treatment, neutralization by using acids, etc., and significant material, energy, and financial inputs are needed. The re-use of alkali-based solid waste can thus provide substantial benefits and savings, together with the rationalization of material flows.

In this paper, waste solidified alkalis from the glass industry are utilized as primary alkaline activators of ground granulated blast furnace slag, which is selected as a representative of the most commonly used aluminosilicate precursors. The main aim of the presented study is to develop eco-efficient building materials with satisfactory mechanical performance. The functional and environmental criteria are supposed to be met concurrently by a proper utilization of waste materials in order to increase benefits arising from the avoided production. Considering the limited knowledge in the field of application of waste solidified alkalis in alkali-activated materials research, the present paper provides data that may contribute to the readjustment of materials flows, increase the rate of recycling, and at the end greening of the building industry.

2. Materials and methods

2.1. Materials

Ground granulated blast furnace slag (GGBFS, Kotouč Štamberk, Czech Republic), an industrial by-product originated during steel production with a specific surface area of 385 m^2/kg, was selected as a primary precursor used within this study. An ARL 9400 XP (Thermo ARL) sequential WD-XRF spectrometer was used to perform the XRF analyses. It is equipped with a Rh anode end-window X-ray tube type 4 GN fitted with 50 μm Be window. All peak intensity data were collected by the WinXRF software in vacuum. The obtained data were evaluated by the standardless software Uniquant 4. The powder samples were pressed into pellets about 5 mm thick and with a diameter of 40 mm and covered with 4 μm supporting polypropylene film. The time of measurement was about 15 min. The phase composition by X-ray diffraction (XRD). The diffractograms were recorded with the help of Malvern PANalytical Aeris diffractometer equipped with $Co_{K\alpha}$ source operating at 7.5 mA and 40 kV. The incident beam path consisted of iron beta-filter, Soller slits 0.04 rad, and divergence slit 1/2°. The diffracted beam path was equipped with a 9 mm anti-scatter slit and Soller slits 0.04 rad. The used detector was PIXcel1D-Medipix3 with an active length of 5.542°. The data were evaluated by Rietveld refinement performed by Profex software (ver. 4.1.0) (Doebelin and Kleeberg, 2015). The content of the amorphous portion was determined with the help of

Table 1
Chemical composition of the used GGBFS.

Compound	SiO_2	CaO	Al_2O_3	MgO	MnO	K_2O	SO_3	Fe_2O_3	Na_2O	TiO_2	BaO
wt.(%)	39.1	38.8	9.8	8.7	0.9	0.7	0.6	0.5	0.4	0.3	0.1

an added internal standard (20% of ZnO).

The chemical and mineralogical compositions of GGBFS are given in Tables 1 and 2. Apparently, the used GGBFS contained high amounts of SiO_2 and CaO and a satisfactory amount of Al_2O_3. The content of amorphous phase was 81.7%, which made further good prerequisites for the alkaline activation. Among the crystalline substances, certain amounts of akermanite ($Ca_2Mg(Si_2O_7)$), calcite ($CaCO_3$), and quartz (SiO_2) were found. Traces of some other silicate minerals (merwinite $Ca_3Mg(SiO_4)_2$, gehlenite $Ca_2Al(AlSiO_7)$, microcline $KAl(Si_3O_8)$) were observed as well in the XRD pattern, but their amount was below 1%.

Waste solidified alkalis (WSA) produced during cleaning processes in the glass industry (AGC Glass, Czech Republic) were used for alkaline activation of the GGBFS precursor (see Fig. 1). Currently, the disposal of this material requires substantial financial means because it is necessary to treat it as hazardous waste. It is disposed of through neutralization (mostly using acetic acid) by an external company. The WSA were composed mainly of NaOH (about 95%), they contained also sulfates (mirabilite, thenardite), and 1% of tin (see Fig. 2). The sulfates may be viewed as an ettringite formation supportive compounds.

The alkali activated aluminosilicate (AAA) mixtures were prepared on the basis of preliminary tests and literature survey. First, the solid WSA was dissolved in water in proportions corresponding to the mass ratios of 24%, 32%, 40%, 48%, and 56% to study the effect of concentration. Consequently, the prepared WSA solutions were stirred for 240 s with GGBFS and quartz sand (0–4 mm) to obtain a homogenous mixture. A detailed information on the composition of the AAA mixes is given in Table 3.

The samples were cast into 40 mm × 40 mm 160 mm molds and kept in laboratory conditions (21 °C, 45% RH) for 2 days. Then, they were demolded (see Fig. 3) and left in the same conditions for another 26 days.

2.2. Methods for analysis of composition and microstructure

The composition of the studied AAA mixes was determined using the same methods as those used for the characterization of raw materials (see Section 2.1). The microstructure was analyzed by a scanning electron microscope Merlin (Zeiss) equipped with a secondary electron detector operating at an acceleration voltage of 15 kV, probe current of 300–800 pA, and a working distance of 6–18 mm. SEM images were analyzed using the NIS-Elements software (Laboratory Imaging Ltd., Prague, Czech Republic).

The pore size distribution was obtained using Pascal 140 and Pascal 440 (Thermo Scientific) devices. The pressure was gradually increased from 100 kPa up to 400 kPa to force the mercury's penetration into the pores of the studied samples. At the evaluation of experimental data, a circular cross-section of capillaries was considered, whereas the mercury contact angle was assumed to be 130° according to Washburn equation.

2.3. Methods for determination of functional parameters

The bulk density was obtained by measuring the mass and volume of five specimens. Before the measurement, the samples were dried at 85 °C until a steady-state mass was reached. The linear dimensions were measured by a digital caliper. The matrix density was determined with the help of a helium pycnometer Pycnomatic ATC EVO (Thermo Scientific). The total open porosity was calculated using the values of bulk- and matrix density. The compressive and flexural strengths were determined according to the ČSN EN 1015-11 standard. The flexural strength was measured by a three-point bending test on three specimens with the dimensions of 40 mm × 40 mm x 160 mm. Consequently, the compressive strength was measured on the left-over specimens after the flexural strength test.

2.4. Methods of environmental evaluation

The environmental evaluation of the utilization of WSA for alkaline activation of GGBFS was performed according to the EN ISO 14040 and 14044:2006 Standards. One metric ton of a mixture was chosen as the functional unit. The obtained results were further used for the calculation of combined functional-environmental indicators to compare the environmental efficiency of particular mixtures with conventional materials including the Portland cement paste, and alkali-activated materials with commercial activators.

2.4.1. System boundaries

The system boundary is depicted in Fig. 4, considering the cradle-to-gate range. Within the analysis, all major material and energy flows were considered, together with the produced outputs in the form of products and emissions.

2.4.2. Inventory analysis

The life cycle inventory included all material and energy inputs, together with the emission outputs (see Table 4). The obtained data were modified according to the functional unit and included in the Simapro LCA software 8.5 [34]. The data for raw material production, processing, transport emission, and energy production were obtained directly from producers, literature, and the Ecoinvent database v.3.5. The Czech energy grid with a dominant share of coal energy and nuclear power plants was considered.

Considering the WSA as a waste material, no environmental impact accompanied with this input was calculated and only emissions related to the material transport were taken into account.

Table 2
Mineralogical composition of the used GGBFS.

Phase	Formula	Wt. (%)
Amorphous phase	–	81.7
Akermanite	$Ca_2Mg(Si_2O_7)$	12.1
Calcite	$CaCO_3$	4.7
Quartz	SiO_2	1.5

Fig. 1. Picture of waste solidified alkalis from cleaning operations.

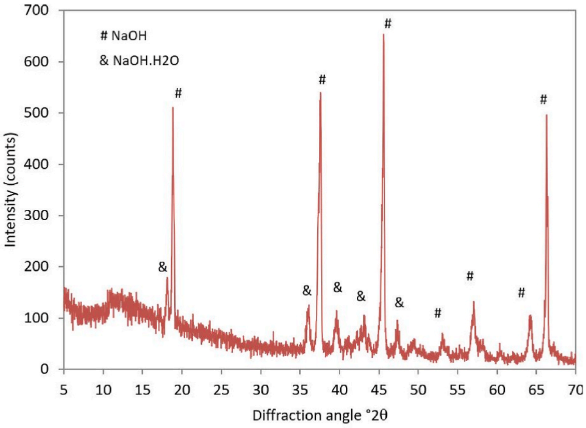

Fig. 2. Diffractogram of WSA

Table 3
Composition of studied AAA mixes (kg/m^3).

Mixture	GGBFS (g)	WSA (g)	Water (g)	Sand (g)
AA1	400	61	252	1200
AA2	400	81	252	1200
AA3	400	101	252	1200
AA4	400	121	252	1200
AA5	400	141	252	1200

2.4.3. Impact assessment

Considering the potential adverse environmental effects, several LCA calculation methods have been introduced to cover sufficiently the potential burden. Among others, CML-IA, ILCD 2011 Midpoint, Impact 2002+, EPS 2015, EPD, EDIP were recognized in several research papers as suitable tools for the determination of environmental effects [35–39]. The main differences usually consist in the level of detail of potential environmental impacts, as well as the capability to distinguish midpoint and endpoint (damage) assessment. While the midpoint indicators can be effectively used for sustainability assessment, endpoint indicators serve rather for comparison purposes. Nevertheless, the above-listed LCA methods usually have very specific characterization factors and usually do not support both approaches. For example, the EPD method has been found as convenient for comparison purposes for endpoint consumers, but, on the other hand, it includes a significant level of aggregation that prevents a detailed understanding of potential environmental externalities. In this regard, the Impact 2002 + methodology (version 3.5) by Simapro 8.5 was used for the calculation of midpoint and endpoint indicators as it allows the combination of both approaches, as well as weighting and single score calculation. This methodology was chosen also because of its suitability for the European region. In addition, the complex list of 15 impact categories provided a robust platform for the identification of environmental burden on both midpoint and endpoint levels [40]. The

Fig. 3. Photo of AAA samples.

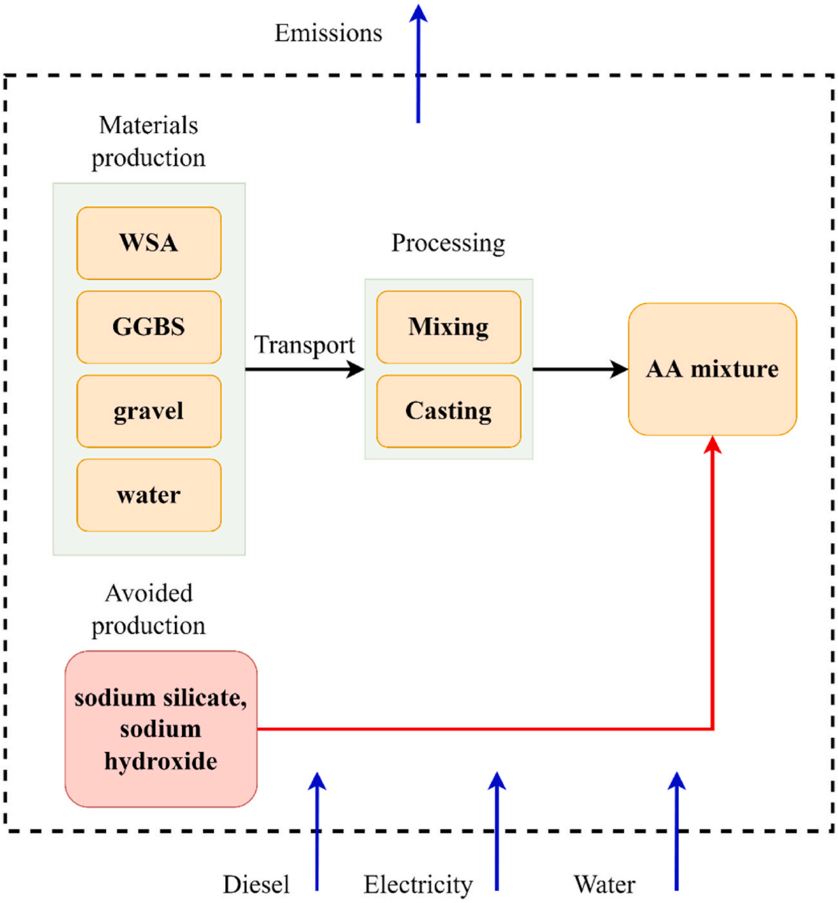

Fig. 4. System boundary.

impact categories included in the assessment were as follows: Aquatic acidification, Aquatic ecotoxicity, Aquatic eutrophication, Carcinogens, Global warming, Ionizing radiation, Land occupation, Mineral extraction, Non-carcinogens, Non-renewable energy, Ozone layer depletion, Photochemical oxidation, Respiratory effects, Terrestrial acidification/nitrification, and Terrestrial ecotoxicity. The calculated midpoint categories were consequently transformed into endpoint indicators (human health, ecosystem quality, climate change, and resources) and used for comparison analysis. The obtained results were used for the calculation of combined functional/environmental indicators for a more explicit interpretation [41].

Table 4
Inventory analysis.

Input	Dataset
Concrete	Concrete 25 MPA (GLO) market for, APOS S
GGBFS	Ground granulated blast furnace slag (GLO)| production
Sodium silicate	Sodium silicate, without water, in 37% solution state (RER)
Sodium hydroxide	Sodium hydroxide, without water, in 50% solution state (GLO)
Water	Tap water (GLO)| market group for
Gravel	Gravel, crushed (GLO)| market for gravel, crushed
Transport	Transport, freight, lorry 16–32 metric ton, EURO4 (RER)| transport, freight
Electricity	Electricity, medium voltage (RER)| market for

2.4.4. Avoided production

The benefits raised from the avoided production of cement or, alternatively, commercial alkaline activators. In this regard, the substitution level of original materials was calculated on the basis of functional properties and the presumed market perception ratio [42]. The material quality expressed by the functional parameters can be compared with the conventional available cement-based products relatively easily [43]. On the other hand, the estimation of market acceptance represents a very difficult task. The disposal of waste alkalis, labeled as hazardous waste, requires the utilization of neutralization solutions (mostly acetic acid) and further processing [44,45]. The re-use of waste alkalis allows to overcome this problem by inducing open material loops and reducing negative impacts. The datasets used for the estimation of avoided production are given in Table 5.

2.4.5. Transportation

The transportation distances may pose a hidden environmental load that should be included in the determination of total environmental impact. A lorry with a loading capacity of 23 t was used for the calculation. The transport of 100 km was considered for both alternatives of commercial activators, and 150 km transport was estimated in the case of waste activator use.

2.4.6. Sensitivity analysis

The sensitivity analysis was carried out in order to assess the impact of avoided production in particular, as the variation in transportation distances does not contribute significantly in terms of summarized outputs. The effects of avoided production of acetic acid only or sodium hydroxide only, and of a complete neglection of avoided production were considered. These alternatives represent major parameters of the performed LCA analysis that may result in a significant uncertainty of the presented outputs.

3. Results and discussion

3.1. Composition and microstructure

The mineralogical composition of the AAA mixtures is presented in Table 6, together with the diffractogram depicted in Fig. 5.

An interesting finding of the XRD analysis was that when the solutions with a very high concentration were used, i.e., in the samples marked as AA4 and AA5, two sodium carbonate minerals, namely Termonatrite and Natron, crystallized. These carbonates were products of the saturation of sodium hydroxide with NaOH by atmospheric carbon dioxide CO_2. The process of formation of these carbonates was caused by the excessive amounts of NaOH in the alkali-activated system. As the mentioned carbonates are obviously soluble in water, their appearance is generally highly undesirable in building materials because, among other things, they can cause efflorescence.

The scanning electron microscopy (SEM) images in Fig. 6A–D revealed that AA1 (activated by 24% WSA solution) had, in general, the finest microstructure among the studied mixtures but microcracks of ~15 μm width were observed, much as in a relatively low amount. In contrast, the AA4 sample activated with 56% WSA solution exhibited a rougher microstructure with more microcracks on the surface, the width of which was though smaller, ~3 μm. The SEM-provided findings on the AA2 and AA3 samples were somewhere in between these two limits.

The pore size distribution curves in Fig. 7 showed that the AA1 mix had the highest amount of pores in the range of 0.01–0.1 μm, while for AA4 it was in the 0.1–1 μm and 1–10 μm ranges. This corresponded with the SEM images in Fig. 6.

3.2. Functional properties

The bulk density, matrix density, and total open porosity are presented in Table 7. The decrease of matrix density was correlated with the increased concentration of the activation solution. As the bulk density was changed only little, the total open porosity decreased from an initial ~28% to ~25%. A comparison with the results of pore size distribution measurement in Fig. 5 showed that the highest total open porosity of AA1 could be attributed mainly to its highest amount of very small pores up to 0.1 μm.

Table 5
Inventory analysis of avoided products.

Avoided production of WSA use	
Sodium hydroxide	Sodium hydroxide, without water, in 50% solution state (GLO)
Acetic acid	Acetic acid, without water, in 98% solution state (GLO)

Table 6
Mineralogical composition of studied materials.

Compound	Chemical formula	Content (%)					
		GGBFS	AA1	AA2	AA3	AA4	AA5
Amorphous phase	–	81.70	80.2	80.3	80.1	68.3	67.2
Akermanite	$Ca_2Mg(Si_2O_7)$	12.10	13.6	12.6	12.4	11.9	10.7
Calcite	$CaCO_3$	4.70	2.5	2.2	1.2	2.8	1.3
Quartz	SiO_2	1.50	0.9	1.2	1.6	1.5	0.9
Hydrotalcite	$Mg_6Al_2(CO_3)(OH)_{16}{\cdot}4H_2O$	–	1.4	1.4	1.7	1.5	2.3
Strätlingite	$Ca_2Al_2SiO_7 \cdot 8H_2O$	–	0.4	0.5	0.2	0.5	0.2
Dawsonite	$NaAl(CO_3)(OH)_2$	–	1.0	1.8	2.8	0.4	3.4
Thermonatrite	$Na_2CO_3{\cdot}H_2O$	–	–	–	–	8.0	8.1
Natron	$Na_2CO_3{\cdot}10H_2O$	–	–	–	–	5.1	5.9

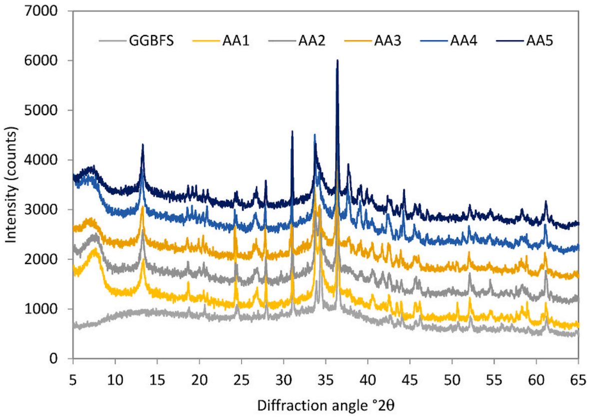

Fig. 5. Diffractogram of studied materials.

The 28-days compressive strength (Fig. 8) showed a decreasing trend with the increasing WSA concentration, but the differences were not very high; all the analyzed mixes remained in a 20–25 MPa range. On the other hand, the flexural strength (Fig. 9) did not exhibit any clear trend; its values were within a 5–7 MPa range. The increase of total open porosity with the decreasing concentration of the activator (Table 7) was thus compensated by the refinement of microstructure (Figs. 6 and 7). However, taking into account the composition (Table 6), only the mixes with the WSA concentration up to 40% (AA1, AA2, and AA3) could be considered as suitable for an application in building practice.

Comparing the obtained results with those achieved by other investigators for commercial NaOH as the primary activator, the compressive strength of AAA mixes analyzed in this paper was somewhat lower than reported, e.g., by Ref. [46]. On the other hand, the results were satisfactory in terms of low-grade applications, or as a replacement of the widely used C20/25 concrete. It should be noted that in case of incorporation of additional sources of silica (e.g., waste glass, rice husk ash) the strength parameters may increase due to the beneficial effect of increased Si/Al ratio on the condensation reaction [47,48].

3.3. Environmental analysis

Fig. 10 shows that substantial environmental benefits may arise in the case of replacing commercially purchased alkaline activators by waste industrial products. The disposal treatment of WSA requires further material and energy inputs that can be, in this case, avoided. In this regard, the utilization of waste activator results in positive outcomes in terms of environmental impact in most midpoint impact categories. This effect is more pronounced in the case of mixtures with WSA concentration 40% and higher, i.e. AA3, AA4, and AA5 (see Fig. 11). The negative environmental impact can be observed in the case of mineral extraction, land occupation, and terrestrial ecotoxicity associated primarily with the GGBFS production. Taking into account the midpoint indicator of climate change expressed in CO_{2eq} kg, all mixtures except AA1 achieved negative values ranging from 15.6 kg to 82.2 kg of preserved CO_2 production. Therefore, such material performance may substantially contribute to greening of the construction industry. The avoided production of commercial NaOH and neutralization agent used for waste alkalis disposal represented substantial benefits in all monitored indicators. Except for a few categories, a majority of negative environmental externalities was compensated in this regard. This finding may be of particular importance, considering the distant statements of various authors, who criticized too narrow criteria focused predominantly on CO_2 emissions only [16].

The cumulative damage assessment presented in Table 8 for each mixture refers to a beneficial impact on human health, climate change, and primary resources consumption. The adverse consequences are related only to the ecosystem quality. The summarized single scored data for each studied mixture are plotted in Fig. 12, where the overall benefits are apparent. Compared to previously published results, a very important improvement was noted for the human health damage category as a substantial drawback

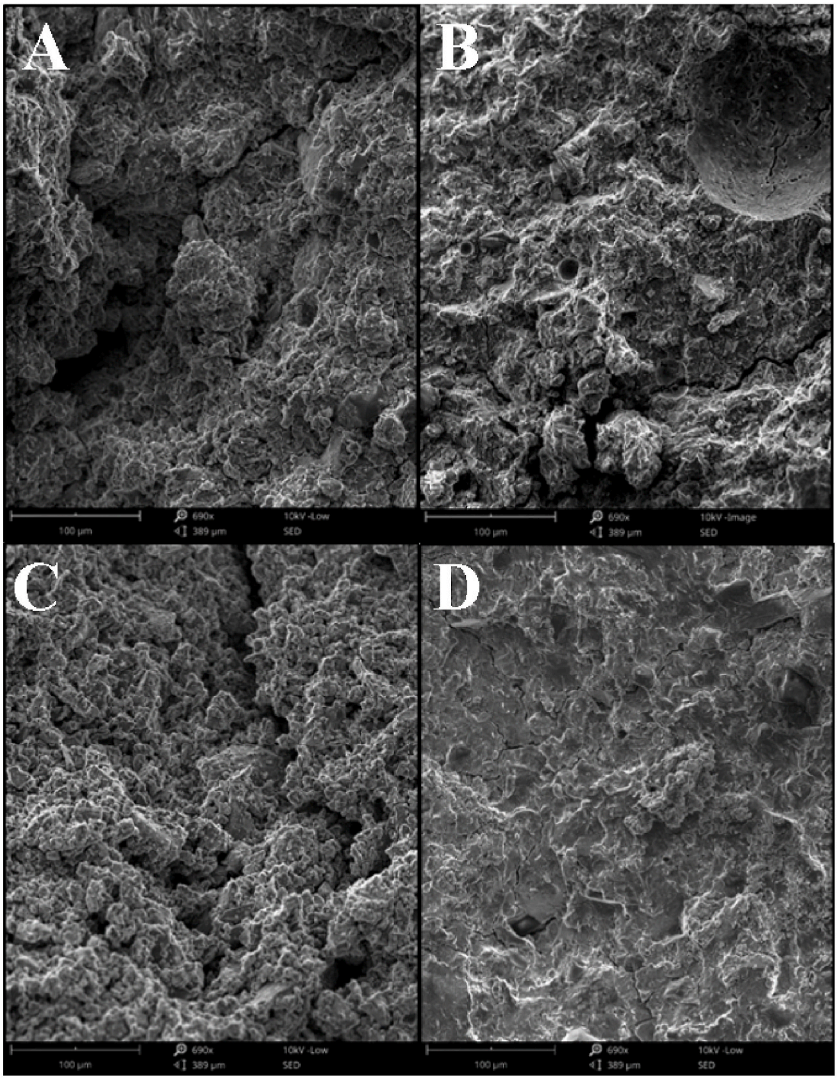

Fig. 6. A - AA1 mixture, B – AA2 mixture, C – AA3 mixture, D – AA4 mixture.

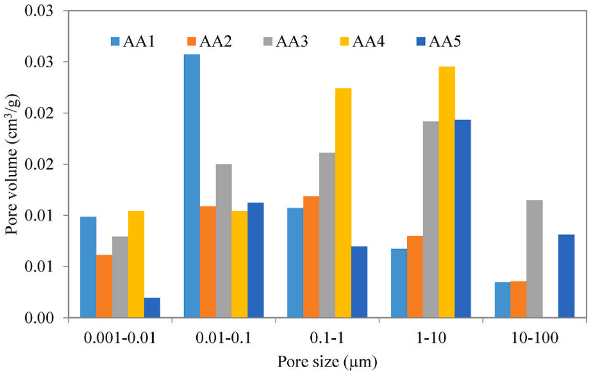

Fig. 7. Pore size distribution of studied materials.

Table 7
Basic material properties.

Mixture	Bulk density (kg/m^3)	Matrix density (kg/m^3)	Total open porosity (%)
AA1	1923	2669	27.95
AA2	1925	2653	27.44
AA3	1939	2658	27.05
AA4	1907	2579	26.06
AA5	1937	2581	24.95

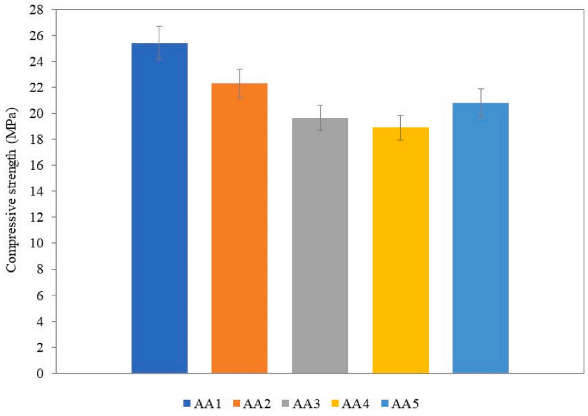

Fig. 8. 28-days compressive strength of studied materials.

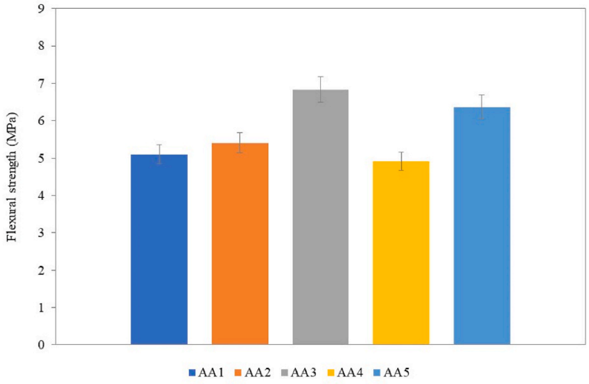

Fig. 9. 28-days flexural strength of studied materials.

accompanied by alkali-activated materials produced from commercial components [28]. This issue was noted in past by several authors, who tried to use alternative activators to reduce the environmental footprint related to commercial activators.

As the WSA used in this study is assumed to be a waste product associated with the consumption of acetic acid for the neutralization on the baseline scenario, a sensitivity analysis was performed to determine the allocation effect of avoided production on the results. As shown in Fig. 13, the benefits of the avoided production represent a substantial issue in terms of environmental burden. On the other hand, the results point at a substantial sensitivity of the presented results to the considered material flows. This assumption could be important if an alternative scenario of waste alkalis disposal (for example long-term storage) is applied or alkali-activated materials manufacturing is not taken into account. In the case when no avoided production was intended, the overall environmental score varied from 25.3 mPt to 27.6 mPt. The effect of transportation distance was excluded from the sensitivity analysis due to its minor effect on final results in a comparison with the avoided production [28].

The obtained environmental score was compared (Fig. 12) with three alternatives: low-grade concrete C20/25, ground granulated blast furnace slag activated by commercial NaOH [29] and sodium silicate [19]. These products were selected to illustrate nominal differences in environmental impacts, and also to understand the negative externalities associated with the use of commercial activators. For the clarity of presentation, only the AA5 mix is presented in Fig. 14. It should be noted in that respect, that the AA5 mixture had the highest WSA concentration, therefore the benefits of avoided production were maximized. Apparently, the global warming potential impact category expressed in kg CO_{2eq} was highest for concrete production. However, in almost all remaining categories the

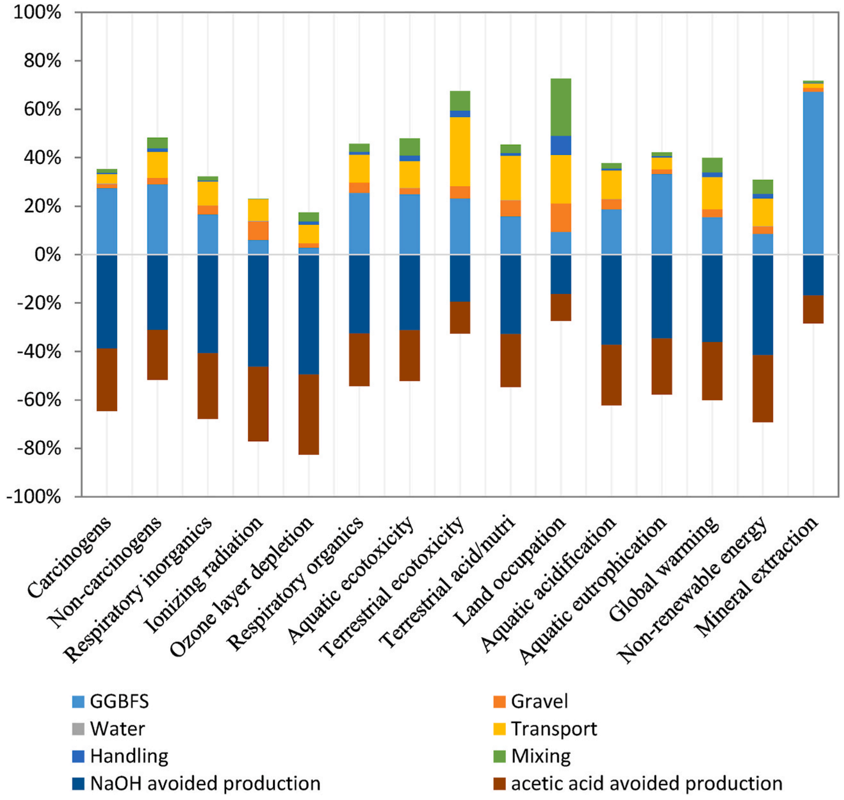

Fig. 10. LCA analysis of AA3 mixture.

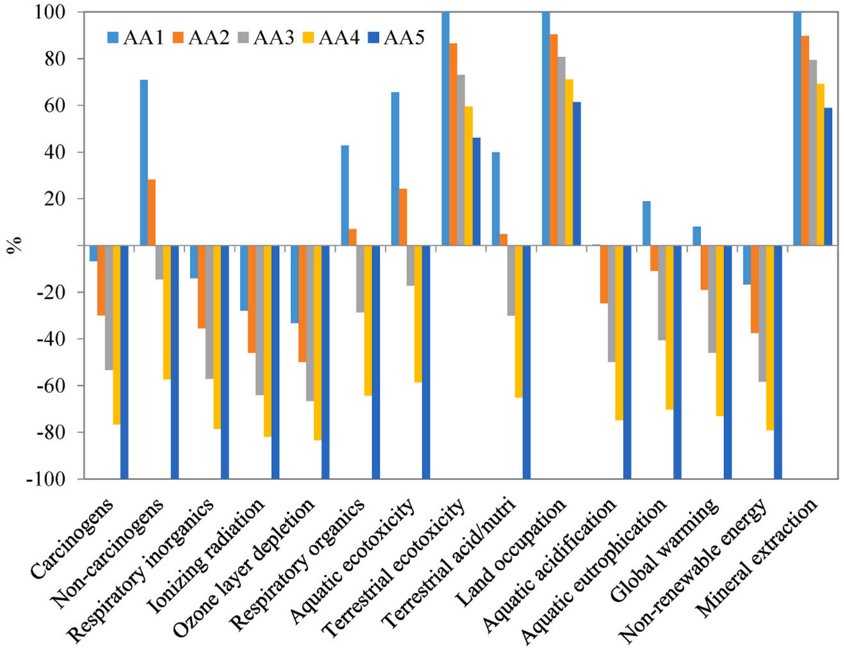

Fig. 11. Comparison of LCA results of studied mixtures.

Table 8
Damage assessment per area of protection.

Damage category	Unit	AA1	AA2	AA3	AA4	AA5
Human health	DALY	−1.44E-05	−4.19E-05	−6.94E-05	−9.69E-05	−1.24E-04
Ecosystem quality	PDF*m^2*yr	28.89	24.70	20.51	16.32	12.13
Climate change	kg CO$_2$ eq	6.63	−15.57	−37.77	−59.97	−82.17
Resources	MJ primary	−376.48	−862.06	−1347.64	−1833.21	−2318.79

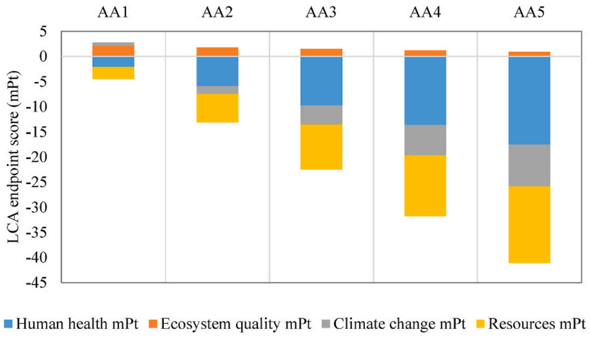

Fig. 12. Endpoint LCA results.

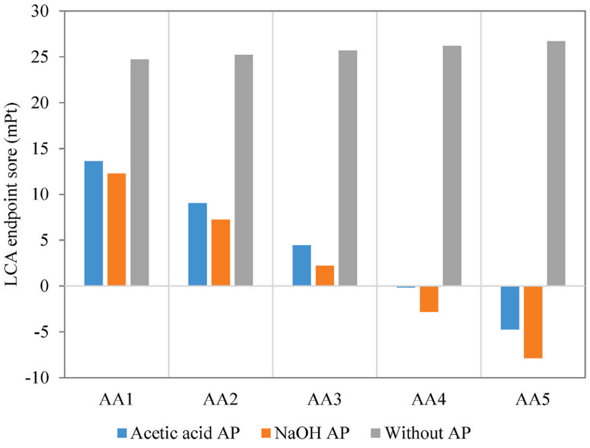

Fig. 13. Sensitivity analysis.

impact of concrete production was not the worst among the compared materials. The GGBFS activated by commercial NaOH presented clearly the highest environmental burden. The GGBFS activated by sodium silicate showed somewhat lower values, except for the mineral extraction. On the other hand, the AA5 mixture based on the utilization of WSA, which was designed in this study, exhibited even negative values, i.e., it was possible to assume a positive impact on the environment, thanks to the avoided production of commercial activator and neutralizing agent. The calculated single score endpoint impact data (Fig. 15) were in a good qualitative agreement with the LCA comparison analysis at midpoint level given in Fig. 14.

Table 9 shows the results of the combined assessment of functional and environmental performance of the AAA mixtures studied in this paper, as compared with the low-grade concrete C20/25 and the ground granulated blast furnace slags activated by commercial NaOH and sodium silicate. The endpoint single score data were used for the calculation of environmental efficiency per MPa as a functional unit of compressive strength in this comparison. The biggest advantage of this indicator is the relatively easy and clearly tangible interpretation, which allows the comparison of materials' efficiency on the basis of just one score. This represents a significant benefit compared to the results of LCA analysis, which are difficult to understand for inexperienced users. The proposed indicator can thus serve as an easy-to-use decision-support tool. Furthermore, the possible asymmetry in the particular functional parameter is taken into account due to the recalculation of the environmental score per unit of the functional parameter. An alternation to the selected approach can be seen in the utilization of the Environmental Product Declaration (EPD) calculation method [49], which provides an easy comparison of particular products or services based usually on four impact categories and, on the other hand, represents a more complex approach compared to relatively narrowly focused methods (carbon footprint, embodied energy). It should be noted, that eco-labeling through the LCA analysis of building materials became a very important issue that will be further developed in the near

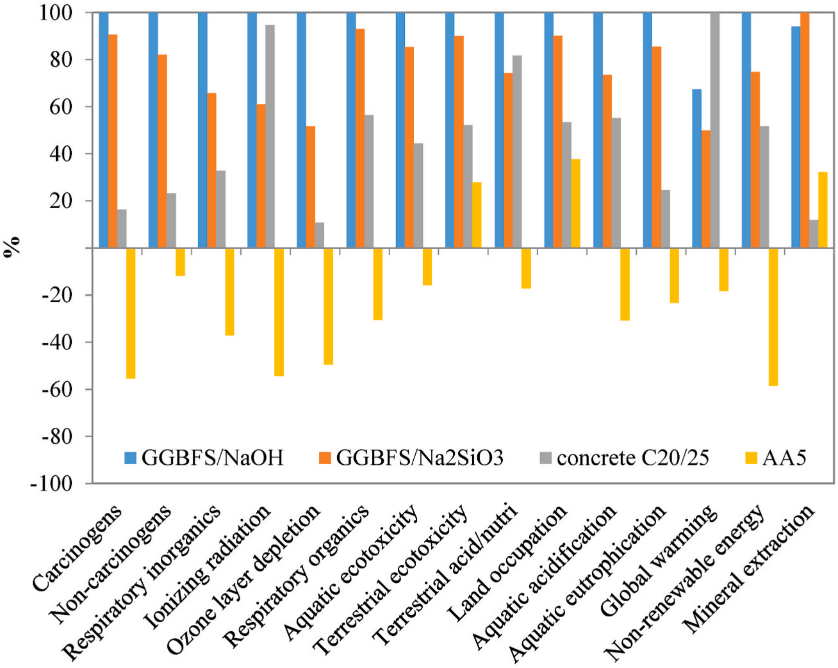

Fig. 14. Results of LCA comparison analysis at midpoint level.

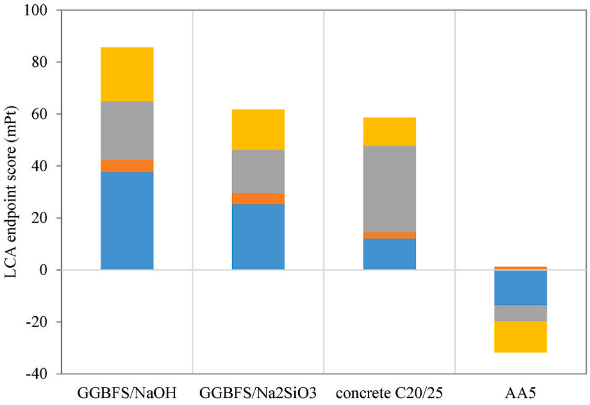

Fig. 15. Results of LCA comparison analysis at endpoint level.

Table 9
Combined assessment of functional and environmental performance.

Mix	Ef (mPt/MPa)	Ef (mPt/MPa) avoided production excluded
AA1	−0.06	1.03
AA2	−0.51	0.88
AA3	−0.92	0.8
AA4	−1.12	0.82
AA5	−1.35	0.93
GGBFS/NaOH	2.95	–
GGBFS/Na_2SiO_3	0.82	–
Concrete C20/25	2.34	–

future to emphasize the principles of rational waste materials management, preservation of natural resources, and mitigation of environmental burden [50].

Considering the obtained results, one can see that the best performance exhibited the AA5 mix with −1.35 mPt/MPa (Table 9). On the other hand, the worst performance belongs to GGBFS/NaOH (2.95 mPt/MPa) followed by concrete C20/25 (2.34 mPt/MPa). A favorable score was achieved by the GGBFS/Na_2SiO_3 mix (0.82 mPt/MPa) since the high compressive strength compensated for the negative environmental impact caused by the Na_2SiO_3 production. The complexity of the proposed indicator, which makes it possible to compare reasonably even materials with very different functional parameters, can be illustrated well when the avoided production would be (artificially) excluded from the analysis scope (Table 9). In such a case the materials designed in this study would achieve similar scores to GGBFS/Na_2SiO_3.

4. Conclusions

The potential of using industrial waste products as alkaline activators of aluminosilicate precursors was analyzed in the paper. Waste solidified alkalis (WSA) from the glass industry were used for the activation of ground granulated blast furnace slag, and functional and environmental parameters of the prepared alkali activated aluminosilicates (AAA) were assessed. The main findings can be summarized as follows:

- The variations in WSA concentration caused only minor changes in mechanical parameters of the studied AAA. All analyzed mixes remained in the 20–25 MPa range for compressive strength and 5–7 MPa range for flexural strength. These values corresponded with construction concrete C20/25 and could thus be considered as satisfactory.
- The observed trends in mechanical parameters were in accordance with the SEM and MIP experiments performed. The increase of total open porosity with decreasing concentration of the activator was compensated by the refinement of microstructure. The XRD analysis of phase composition though revealed that only the mixes with WSA concentrations up to 40% would be suitable for an application in building practice; the use of higher WSA concentrations resulted in the appearance of soluble carbonates in hardened AAA mixes which could cause efflorescence.
- The environmental analysis showed a significantly reduced environmental impact of the designed AAA activated by WSA, as compared with construction concrete C20/25 and GGBFS activated by commercial NaOH and sodium silicate. The benefits associated with the avoided production of acetic acid used for waste alkalis neutralization and with the NaOH production were identified as the main reasons.
- The combined assessment of functional and environmental parameters revealed a somewhat lowered environmental impact of the analyzed AAA even in the case of neglecting the avoided production, as documented by determining the endpoint single score data per 1 MPa as a functional unit of compressive strength. In the case of inclusion of the avoided production, all studied mixes exhibited very positive effects on the natural environment. The applied method of AAA design thus showed a very prospective path to further greening of the construction industry.

As it was shown in this study, the interpretation of results presents an important task of sustainable materials design. Besides the detailed identification of relevant environmental hazard impact categories, the environmental assessment requires also the comparability of the obtained results with environmental profiles of other materials, in the most desired way as a decision-supporting tool that incorporates the functional performance. The proposed method can be very useful in terms of merging the functional and environmental indicators for the maximization of environmental/functional efficiency.

Author statement

Jan Fořt: Conceptualization, Methodology, Data curation, Supervision, Funding acquisition, Writing- Original draft preparation Martin Mildner: Investigation, Data curation, Martin Keppert: Visualization, Investigation. Robert Černý: Writing- Reviewing and Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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3.5 Sustainable building materials' design using interdisciplinary research

Selected journal paper

Fort, J., Cerny, R., 2022. Limited interdisciplinary knowledge transfer as a missing link for sustainable building retrofits in the residential sector. *Journal of Cleaner Production 343. 131079.*

Despite the numerous researchers aimed at the development of innovative eco-friendly building materials, the improvement is rather minor and faces several barriers. It is necessary to identify the major issues that prevent the wider acceptance of sustainable measures in the construction industry. On this account, the development of building materials cannot be deemed as a technical discipline only, and knowledge and assessment methods from various relevant scientific disciplines need to be involved. As sustainability stands on technical, economic, social, and environmental pillars, the recent advances from these fields may be a key to its success. In this section, a critical review summarizes the main issues that prevent the faster adoption of sustainable measures and provides the context of the necessity of addressing the major barriers to relevant stakeholders.

Contribution to the practice: As shown in this study, the successful adoption of sustainable principles requires more interconnected cooperation and intensified transfer of recent knowledge between particular scientific disciplines. In other words, the challenges associated with the construction industry cannot be accepted as a technical discipline only, thus the creation of new communication platforms for knowledge exchange and sharing is required.

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Limited interdisciplinary knowledge transfer as a missing link for sustainable building retrofits in the residential sector

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ABSTRACT

Despite the substantial effort on almost all levels during the last decades, the buildings' renovation rate needs to be at least doubled from the current 1% to meet ambitious energy efficiency goals. In the same way, the energy-intensive material replacement did not reach yet the desired grade in terms of sustainability measures and outlined goals heading to a low-carbon economy. This paper summarizes principles of sustainable development together with the current methodological framework relevant to the civil engineering and construction industry. The main part is devoted to the identification and understanding of principal factors preventing faster adoption of energy efficiency measures. High initial financial costs, investment risk over a long-term period, poor acceptance of sustainable measures, lack of information, and limited methodological framework for a reliable evaluation of environmental projects with the intergeneration context or understanding of externalities of human activities are identified as the main barriers to the sustainable building retrofits. These barriers are closely related each other and can be merged into several groups according to the stakeholders or scientific disciplines to amplify their primary impact. Notwithstanding, solution strategies based on narrow boundary conditions and limited multidisciplinary approach prevent substantial advances towards the sustainable building sector. In this regard, the major obstacles preventing the achievement of energy efficiency goals can be remedied by interdisciplinary cooperation.

1. Introduction

The significantly higher temperatures above the Arctic Circle, droughts and floods in Central and Eastern Europe, devastating storms at the western coast of Europe are responsible for many human lives and massive devastation of biodiversity. From the financial point of view, the weather-related events caused damages reaching € 240 billion in 2017 and more irreversible changes will affect a substantial part of the European population for several upcoming decades in particular (Ricke et al., 2018). The current trend of temperature increase poses a considerable risk for future life and economic stability and the development of humankind. Since the concerns associated with the negative externalities of human activity and population boom on global climate are increasing, various action plans to mitigate this threat take place (Kober et al., 2020; Biskaborn et al., 2019) (see Table 1).

Since the negative human activity footprint was distinguished as a considerable threat for future generations and world economic development, the term sustainability became more relevant and sustainability measures were used as a general framework for the development of specific measures and indicators. Despite the definition of basic pillars of sustainability, primary concerns are related to the depletion of natural resources and thus adverse affection of the ecosystems (land and sea surface area) by human activities (Kawashima et al., 2019; Fort et al., 2020). Costs related to the manufacturing of raw materials and necessary expenses for further disposal create, together with maintenance costs and the investment return, a substantial part of the economic pillar (Nong et al., 2020; Marti-Ballester, 2019). The framework of economic concerns also deals with the vulnerability of product chains to the instability of the prices and possible impacts on geopolitical issues (Kaviani et al., 2020; Singhal et al., 2020). The social level of sustainability aims at fostering people towards the development of better quality of life by the improvement of human health, health care, living conditions, security, or working conditions (Sharpton et al., 2020).

In order to meet the sustainability criteria, the European Union committed itself as a leader in global climate action and reaching net-zero greenhouse gases (GHG) emission targets until 2050 (Bandeiras et al., 2020; Say et al., 2018). Such an ambitious target related to holding temperature increase below 2 °C requires a coherent strategy for the modernization of the European industry and economy as introduced

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List of abbreviations

EPBD	Energy Performance of Building Directive
LCA	Life Cycle Assessment
LCEA	Life Cycle energy Assessment
EPD	Environmental Product Declaration
GHG	Greenhouse Gas
LCC	Life Cycle Cost
LCI	Life Cycle Inventory
GDP	Gross Domestic Product
NPV	Net Present Value
EPS	Environmental Priority Strategies
EVR	Environmental-Costs/Value Ratio

Table 1
Literature selection process.

	Parameter	Condition
1	Time period	From 2000 to 2021
2	Subject area	Environmental Sciences, Engineering Environmental, Green Sustainable Science and Technology, Construction and Building Technology, Engineering Multidisciplinary, Engineering Civil, Economics, Social Sciences, economics, Econometrics
3	Publication type	Research articles, reviews
4	Language	English
5	Full-text availability	Available on-line as full-text

in the European Commission's strategic long-term vision for 2050 – A Clean Planet for All (Gottinger et al., 2020). As widely accepted, the building sector is considered very energy-intensive, even compared to the transportation or industry in the European Union, since it is responsible for ~40% of the total energy production and ~36% of carbon dioxide emissions (Li et al., 2019a; Filippidou et al., 2017). In other words, despite many initiatives aimed at the minimization of environmental footprint, the building sector still has the greatest potential for energy efficiency improvements since a very significant part of current building stock was built in the previous century when the requirements for energy efficiency were lower. Given today's possibilities in terms of materials used, insulation, or heating methods, together with the fact that the building sector is the most energy-intensive sector, substantial improvements can be achieved (Gottinger et al., 2020; Sesana and Salvalai, 2018). Nevertheless, the energy efficiency of the current building stock in the EU does not meet the modern criteria, and more than 75% of the buildings are inefficient (Sesana and Salvalai, 2018). The incurred pressure on the primary energy production results in the increasing need for sustainable building retrofits (Bandeiras et al., 2020; Elkhapery et al., 2021).

In light of recent studies, the operation stage of buildings presents the most energy-demanding phase during the whole life cycle (Dewulf et al., 2009). In order to decrease this undisputable harmful effect of the building sector, all EU members created a framework regarding the transition to a low-carbon future of the building sector (Ascione et al., 2016). The adopted EU Directive (2010/31/EU) highlighted the role of institutional building leaders in the field of energy efficiency and substituted the previous 2002/91/EU Directive (Joensuu et al., 2020). The principal purpose of the mentioned EU Directive lays especially in necessary actions for the reduction of energy demands for heating and cooling. Furthermore, the Energy Performance of Buildings Directive (EPBD) sharply defines "nearly zero energy buildings" to deliver guarantees for sustainable buildings with high-performance thermal stability and decreased energy demand based mainly on renewable energy sources (Kalaycioglu and Yilmaz, 2017). Such effort resulted in requirements for the new public buildings which shall fulfill the nearly zero energy standard from 2019 and from 2021 all new buildings are supposed to match this energy efficiency criterion. In other words, the sustainability measures became very important in terms of building stock planning (Joensuu et al., 2020) as a broadly accepted term heading towards conservation and development of humankind for the positive future perspectives (Adabre et al., 2020; Bahramian and Yetilmezsoy, 2020). However, there are obviously missing links between the individual sustainability pillars which should be investigated, defined, and cleared (Hasan et al., 2020).

On top of the three main areas of concern, it is possible to identify also the fourth one, the technical concerns (Dewulf et al., 2015). Reaching sustainability has to be connected with relevant technological alternatives in terms of physical resources quality, availability, renewability, and other criteria that need to be supported by sufficient political will and commitment (Fig. 1). The insufficient technical solution without alternatives to natural resources can be viewed as a substantial barrier to sustainability (Jia et al., 2018). Considering the energy consumption of the building industry, it is possible to recognize gradual solutions for particular obstacles. In general, with the reflection of the sustainability pillars, these barriers were the subject of interest of scientists, policy makers, building inhabitants and other stakeholders during the previous decades, and especially in the present when the needs for the achievement of sustainability are even higher (Illahi and Mir, 2020; Martek et al., 2019). The decomposition of the major problem dealing with the poor renovation rate of the current building stock can be a prospective way towards the improvement of the renovation process from the viability point of view (Reuter et al., 2019).

This paper describes the current state-of-the-art from the perspectives of individual sustainability pillars to identify the main barriers to the fulfillment of previously described targets. The major constraints are accompanied by the division of the identified obstacles into isolated problems, which are often solved independently of each other, although they are very significantly interconnected according to individual

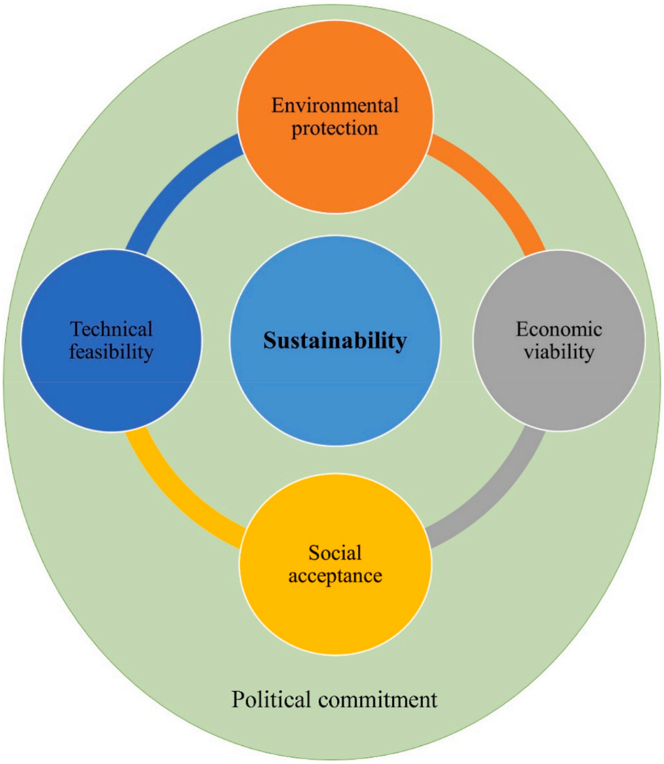

Fig. 1. Extended interpretation of sustainability pillars.

stakeholders, scientific disciplines, etc. In these terms, the issue of low renovation rate of current building stock does not rely on any hidden barrier, not described in previously performed studies but rather on the isolated solution of particular obstacles with only minor overlap to other areas. This is associated with a limited transfer of knowledge and advanced methods developed in recent years, which does not sufficiently synchronize individual activities and limited relevance. Last but not least, it is necessary to mention that the issue of the building sector transition cannot be seen as a purely technical solution, but also as a socio-psychological problem viewing technical arguments as a knowledge and support base. The missing link for the achievement of sustainable measures in the building industry thus lies in the neglect of the scale and complexity of the whole process which leads to insufficient sharing of new knowledge and its utilization. Finally, this study demonstrates that a combination of knowledge from several scientific fields is necessary to achieve a synergic effect of the multidisciplinary approach which can significantly contribute to the low energy performance of the building industry and promote the attainment of ambitious carbon dioxide restriction goals in the EU region.

2. Review methodology

In this paper, a systematic literature review was performed to identify the relevant existing studies and provide an overview of the analyzed research field, formulate the research questions, and synthesize the contributions to the current-state-of-the-art. In this regard, all steps required for the reproducible screening strategy of literature sources are provided, as well as the limitations of the study (Caiado et al., 2017). The methods used for the literature review arise from previously published recommendations, such as those formulated by Siddaway et al. (2019).

The bibliometric analysis of the current state-of-the-art was performed according to the Proknow-C method (de Carvalho et al., 2020) as a verified tool to organize knowledge for a literature review which comprises the following steps:

1) Definition of the research questions and primary goal.
2) Elaboration of the bibliographic portfolio - selection of suitable database sources and search based on relevant keywords.
3) Bibliometric analysis - listing results of the conducted search, selection of relevant studies, use of subject filters.
4) Systemic analysis - removal of irrelevant papers, full papers analysis.

2.1. Definition of the primary motivation of the study

The main motivation for this study elaboration arised from the complexity of the building retrofits issue based on the sustainability pillars. The main goal of the research was not focused on the exploration what has been found in available literature dealing with the barriers in the buildings retrofitting only by the meaning of technical feasibility or economic costs directly associated with building retrofitting, but rather on the understanding of driving forces standing in the background, covering barriers and synergies between sustainability pillars. In other words, the main research objectives can be divided into the following research questions:

Q1 – Is there a robust enough knowledge base to achieve sustainability in the building sector?

Q2 – What are the barriers and synergies within the particular pillars in achieving the sustainability goals?

Q3 - Is there sufficient interdisciplinary communication sharing the theoretical concepts and advances in relevant fields?

2.2. Data sources

Google Scholar, Scopus, and Web of Science can be considered as comprehensive databases of science articles (Chapman and Ellinger, 2019). However, Google Scholar has been criticized for listing several predatory journals, thus papers with limited originality and validity can be found there. In this regard, Google Scholar was avoided due to limited quality control and the risk of invalidated outcomes (Aguillo, 2012).

Web of Science was selected as the primary source for the journal articles as the most comprehensive database with high-impact publications. These data were completed by articles from the Scopus database as the largest abstract and citation database worldwide. Based on the recommendation presented in previously published review papers, merging both Web of Science and Scopus data within the literature analysis can be considered as the most sensible approach to include all relevant studies. Journal papers have been completed by relevant reports to provide background data from European institutions.

Data for the study were collected in three consecutive steps comprising the database, supplementary and conclusive search. At the first stage of the literature survey, the following search phrase was used to identify relevant literature:

Sustainability, sustainable development, sustainable barrier, sustainability pillar, building retrofit, building energy, constructions, renovation, energy efficiency, buildings with the Boolean expression “AND”. Used keywords were combined to cover all combinations relevant to the scope of the literature survey. The papers were identified with the help of used keywords in the titles, abstracts, and keywords as well. Within the performed literature search, the following criteria were considered to strengthen the quality of the review and to select the most relevant papers.

Selected papers were sorted according to the research areas to deliver highly relevant studies which contribute to the research scope. Non-English studies were excluded as well as studies without a full-text availability. Consequently, titles and abstracts were reviewed to filter studies in the scope of this study. The list of the pre-selected papers was subjected to in-depth analysis to discard irrelevant studies and to provide a list of relevant studies to the research questions.

In addition to the publication identified by the database search, further literature sources were retrieved to elucidate theoretical concepts in the background together with a discussion of presented results.

To assess the quality of the used references, the impact factors (2020) of journal articles are shown in Fig. 2 along with the number of records per journal. The impact factors of scientific journals are annually published in the Journal Citation Report by the Thompson Reuters/Clarivate as a worldwide known parameter describing the relevance of the scientific outputs.

Considering the regional differences in the legislative and methodological approaches, the geographical distribution of analyzed studies is illustrated in Fig. 3. Since several studies could not be assigned to specific geographic areas, they are listed as “not specified”.

2.3. Limitations

The first limitation of the performed study arises from the use of two scientific databases (Web of Science and Scopus) and used search strings. It is therefore possible that some studies have been overlooked and their findings are not reflected. On the other hand, Web of Science covers the most respectable journals in this field, thus major knowledge should be covered sufficiently.

The high dropout rate (over 95%) of studies from the preliminary search may result in omitting a relevant study, for example, for reasons of using different terminology. In this sense, the consequent review studies should revisit the used search strings and terminology to cover potential gaps.

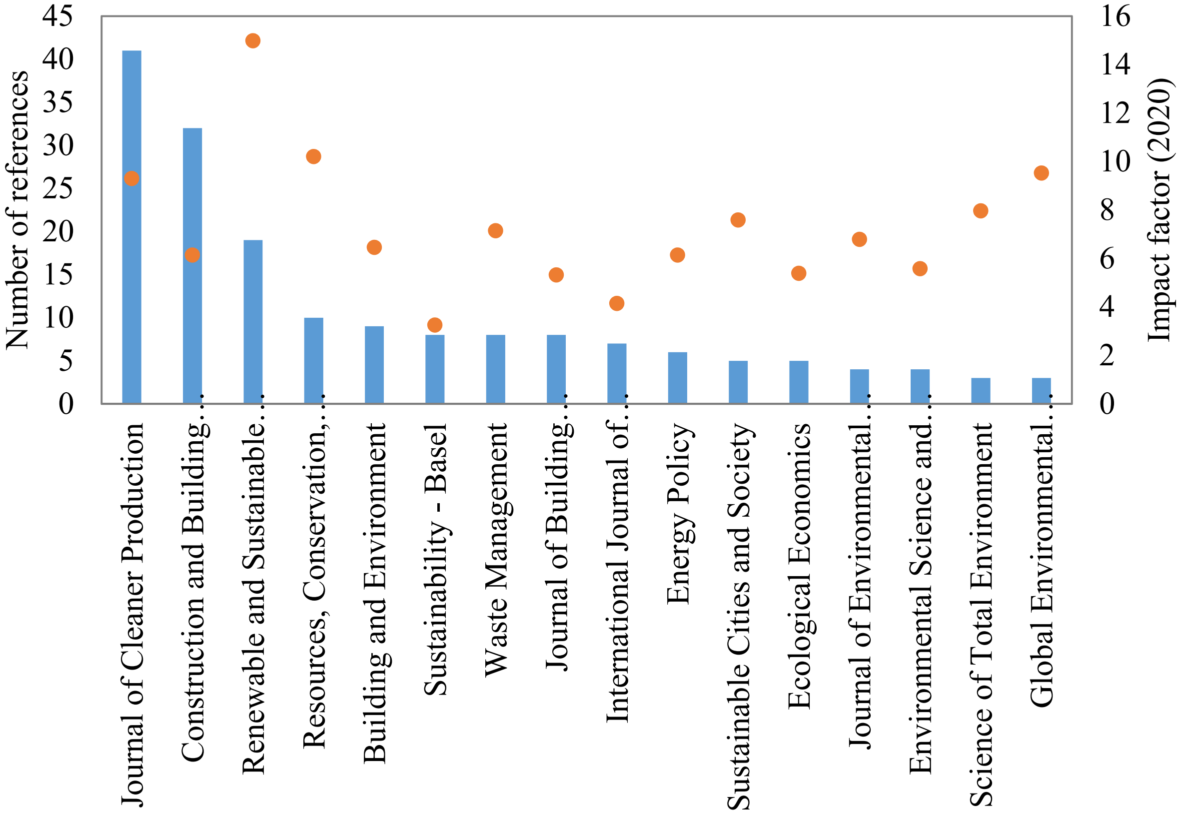

Fig. 2. Impact factors of identified studies.

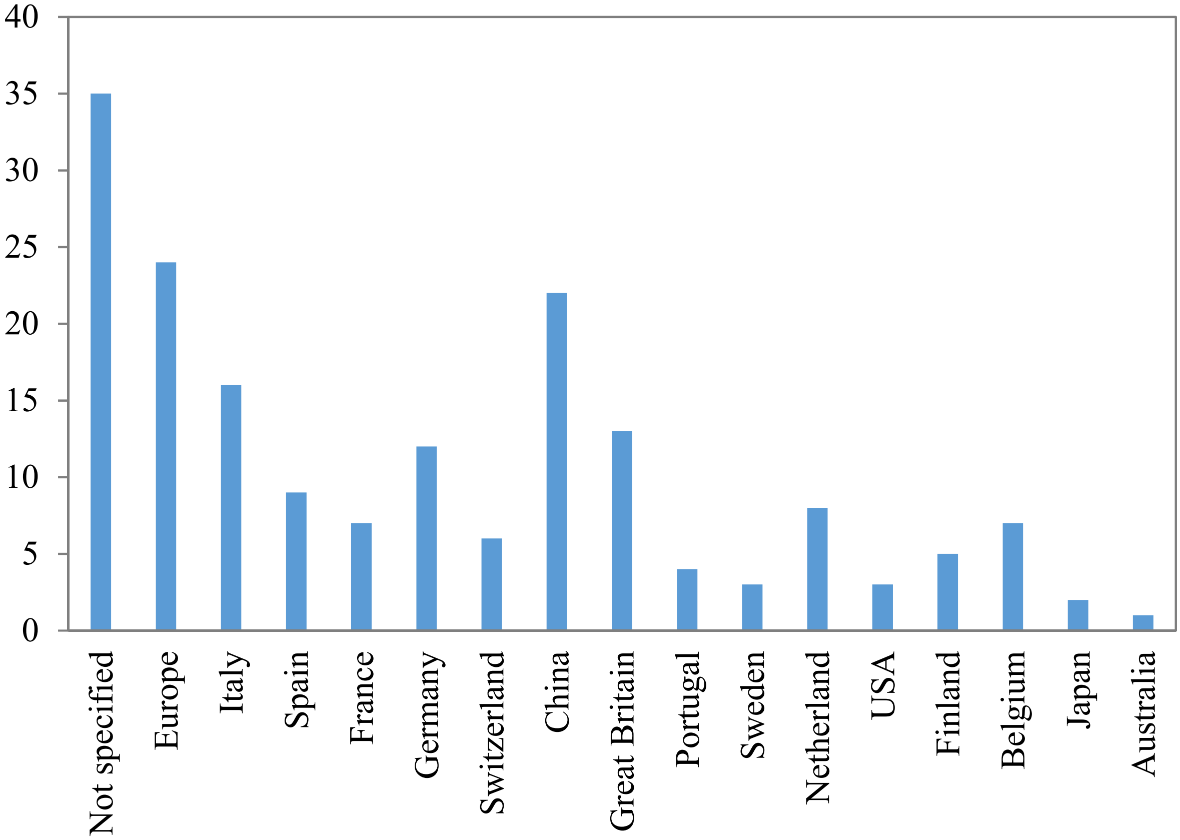

Fig. 3. Country of origin of relevant studies.

3. Sustainability barriers

Considering the understanding of sustainable development based on individual pillars, it is necessary to identify well-described areas within each pillar, as well as areas that are not sufficiently reflected or perceived with sufficient emphasis. In this regard, three major areas corresponding to basic sustainability pillars (Nong et al., 2020) dealing with goals in the building sector were extended by the technical feasibility described in (Dewulf et al., 2015; Martek et al., 2019). The following areas are described in greater detail: environmental implications, technical feasibility, economic viability, and social acceptance.

3.1. Environmental implications

Since the 1970s, environmental issues have been recognized as an important part of the development of human society, and the concept of sustainable development was beginning to be viewed as a prospective way without damaging future generations (Silvestre et al., 2014). The technological advances were supposed to take into account the environmental aspects of human activities and their relations between life quality and possible threats for the future. However, a broader acceptance and expansion of this concept was delayed, except for a few examples, such as the Resource and Environmental Profile Analysis that became known from 1975. However, the integration of sustainable measures does not satisfy the expectations until now. In other words, the balance between economic growth and environmental externalities still represents a challenging task that needs to be resolved (Subramanian et al., 2018). Even facing such important threats as the global climate change undoubtedly is, the lack of standardization of the environmental assessment tools still presents a significant obstacle against a coordinated action (Loiseau et al., 2012). Therefore, appropriate communication of the outcomes of scientific analyses represents an essential element for any future attempts aimed at the removal of sustainability barriers.

3.1.1. Applicability of environmental assessment tools

The most common methods related to environmental assessment should be compared, and discrepancies and ambiguities clarified. Across the literature, it is possible to distinguish many different or only derived methods used for the determination of human and environmental risk assessment. These methods highly vary in their scope, inventories flow, scale, applied indicators, feasibility, and sensitivity on input parameters. Ecological Footprint (Lamnatou et al., 2018; Li and Wen, 2018; Yang et al., 2018), Material Flow Analysis (Stanisavljevic et al., 2015; Makarichi et al., 2018), Substance Flow Analysis (Jensen et al., 2017; Kim et al., 2017; Philis et al., 2018; Pfaff et al., 2018), Input-Output Analysis (Bosch et al., 2015; Nuss et al., 2016; Singh et al., 2017; Soulier et al., 2018), Exergy Analysis (Ghannadzadeh and Sadeqzadeh, 2016; Ali et al., 2018; Tran et al., 2018; Acikkalp et al., 2018), Emergy Analysis (Law et al., 2017; Zhang et al., 2018; Wu et al., 2018; Liu et al., 2019), and Life Cycle Assessment (LCA) as probably the most frequently used (Sim, 2016; Ji et al., 2016; Vieira et al., 2016; Cabeza et al., 2014; Penaloza et al., 2016; Bizjak et al., 2017; Weiler et al., 2017; Vilches et al., 2017; Rock et al., 2018; Hossain and Poon, 2018; Al-Ayish et al., 2018; Chau et al., 2015) can be considered as the main representatives of environmental assessment tools.

Each method has its own strengths and weaknesses. Nevertheless, Loiseau et al. (2012) compared nine different methodologies including the above listed using several criteria. They concluded that LCA can be perceived as the most promising framework for the environmental assessment. The insufficient evaluation of some issues, such as biodiversity, land use, and water use, needs though further improvement and adaptations, as well as the development of methodological instructions for a regional scale. Huysman et al. (2016) then emphasized the cut-off problem related to the limited boundary condition and underestimation of flows outside the boundaries in that respect.

The explanation of the current extensive LCA use can be found in its standardization as early as in 1997, which made possible a broader application of life cycle thinking in the practice. Despite the relatively long experience with LCA for environmental evaluation of production processes, the use of this method in the building industry was not so extensive as in other areas for a relatively long time (Buyle et al., 2013). The discovery of substantial negative environmental consequences, together with the widespread use of primary energy, sharply highlighted the environmental aspects of the building industry and attracted the attention of governments and scientists. The wide range of LCA methods covering environmental evaluation during the whole service life brought this systematic, comprehensive approach to the decision-making process for the optimal selection of particular products (Cabeza et al., 2014). In the current literature, it is possible to identify hundreds of papers dealing with the life cycle assessment of buildings, and well as review papers integrating advances in this field (Jia et al., 2018; Weiler et al., 2017; Buyle et al., 2013; Arcese et al., 2018; Bertone et al., 2018; Edelen et al., 2018; Johnsen and Lokke, 2013; Karimpour et al., 2014; Pieragostini et al., 2012). Many of those studies were aimed either at particular production stages of the building (Zhang and Wang, 2016; Gan et al., 2017; Rodrigues et al., 2018; Coelho and de Brito, 2012; Vitale et al., 2017) or at macro-scale analyses of the building stock (Jia et al., 2018; Huysman et al., 2016; Stephan and Athanassiadis, 2018; Abdallah et al., 2016; Baldwin et al., 2018).

Regardless of the high scientific activity devoted to the estimation of the environmental footprint of the building industry, both research and practice are vastly fragmented as many different LCA tools are used (Haapio and Viitaniemi, 2008). The conclusions of this study aimed at the characterization and classification of frequently used LCA tools are, perhaps surprisingly, still actual at present. The problematic comparison of performed analyses' outcomes was also described later by Cabeza et al. (Cabeza et al., 2014; Mastrucci et al., 2017). The observed difficulties could though be attributed also to different indoor thermal requirements, fractioned legislative framework, and different climatic conditions. The comprehensive work of (Chau et al., 2015) dealt with the assessment of three various approaches or groups of environmental impacts. Namely, Life Cycle Assessment (LCA) and Life Cycle Energy Assessment (LCEA) together with Life Cycle Carbon Emissions Assessment ($LCCO_2A$) as alternatives were subjected to a thorough analysis to compare their data inventories, methodology framework, and boundary scoping, which poses the primary barrier against supporting the decision-making process towards sustainability. However, even a deeper subsequent derivation of such approaches could not bridge the problems with the limited scopes of LCEA or $LCCO_2A$.

Based on LCA, the environmental product declaration (EPD) was defined as a guideline for building construction and civil engineering (Passer et al., 2015). Notwithstanding, a variety of approaches can be distinguished across Europe due to local differences in climate, energy mix composition, and transportation systems. On the other hand, several countries (e.g., Germany, Switzerland, Belgium) already implemented the EPD program into building certification systems as a competitive advantage referring to nearly-zero energy buildings policy. Since the efforts paid to the clarification of results of various impact categories, biodiversity, land use, and toxicity, in particular, took place, calls for the harmonization of environmental performance evaluation became more important (Cabeza et al., 2021; Eberhardt et al., 2021). As noted by Passer et al. (2015), harmonization in these terms needs to be coordinated at a better level to avoid divergent results. Specifically, the available reports in the literature refer to required improvements in the life cycle inventory data as a crucial input for coherent quantification of the environmental footprint.

3.2. Technical feasibility

Understanding the environmental score of building materials production, labeling of building energy efficiency represents only one side of the coin. The technical feasibility in terms of the availability of eco-friendly materials poses another important task that needs to be resolved properly to reach the outlined goals (see Table 2).

During recent years, increased attention was paid to the investigation of less energy-intensive materials to replace Portland cement, natural aggregates, or other components used in the construction phase (Nwankwo et al., 2020; Aydin and Arel, 2019; Memon et al., 2019; Hamada et al., 2018; Meng et al., 2018). The replacement of natural aggregate based on the utilization of various industrial waste products became a very popular way for the minimization of waste cumulation. Various replacement alternatives for both natural aggregate and cement were identified according to the local availability (Table 3). As one can see, cement or aggregate replacement is widely studied in terms of the availability of local by- or waste products based on similar chemical composition and pozzolanic activity. Therefore, the possible utilization of materials such as rice husk ash or palm oil fuel ash cannot be applied in the European context. On top of that, the utilization of coal fly ash will be in the close future significantly reduced due to restrictions on the operation of coal-fired power plants. Considering the material resources available in Europe, viewing the old buildings as a potential resource bank may be of eminent attention due to the fact that it would be possible to create closed material loops and thus substantially rationalize material flows.

However, the natural aggregates replacement strategy does not provide sufficient mitigation of negative environmental impacts since the long transportation routes hinder the overall benefits of alternative materials use (Di Maria et al., 2018a) although 15–20% savings can be expected (Colangelo et al., 2020). Among others, granulated blast furnace slag, fly ash, silica fume, rice husk ash, wood waste ash, or red brick powder were found as prospective alternatives that may serve as partial cement replacement in binders (Nwankwo et al., 2020; Enriquez et al., 2021; Liu et al., 2021a; Bilginer et al., 2020; Hafez et al., 2020; Fort et al., 2019). Substantial attention was paid also to the recycling potential of construction and demolition waste. For example, Pedreno-Rojas et al. (2020) studied the effect of waste gypsum on the workability of plasters. Khan et al. (2020) found the use of waste glass in cement-based materials as an effective solution for both minimizations of natural resources consumption and mitigation of negative consequences of other possibilities of waste glass disposal. Besides the partial cement or natural aggregate replacement, alkali-activated materials became a hot topic for materials engineers due to the possibility to avoid the use of cement completely. However, this emerging field is still lacking the knowledge for broader application despite several real applications (Provis, 2018; Koci et al., 2020; van Deventer JSJProvis and Duxson, 2012; Amran et al., 2020). An intensive effort of many scientists worldwide may allow the application of less-energy intensive binders originated from various abundant waste materials to a greater extent soon and thus decrease the environmental footprint during the construction phase (Liu et al., 2021a; Gavali et al., 2019). As concluded by Hossain et al. (2018), the optimized concrete design based on the utilization of various supplementary cementitious replacements can decrease the global warming potential by about 20–38% depending on the applied methodology and used SCM. More favorable results were shown in the work of Di Maria et al. (Di Maria et al., 2018b), where reduction of global warming potential was achieved by alkaline activation of stainless-steel slags. A substantial reduction in GHGs emissions as a result of using the alkaline activation process was confirmed by other authors as well (Fort et al., 2018; Abdulkareem et al., 2019), but, on the other hand, considerable risks in other impact categories were identified (Fort and Cerny, 2020; Habert et al., 2011).

Table 2
Number of studies.

Search parameter	Number of records	
	Scopus	Web of Science
Keywords	6904	6370
Time period	6352	5872
Subject Areas	4716	4428
Publication type	3526	3016
Language	3512	2946
Full-text only	3065	2374
Relevant to the scope	148	105
Duplicate results	76	
Final record	**177**	

Table 3
Examples of traditional binders/natural aggregates replacement alternatives.

Replacement source	Type of replacement	Reference
Silica fume	Cement	(Burroughs et al., 2020; Gupta and Kua, 2020)
Coal fly ash	Cement	(Lee et al., 2020; Hossain et al., 2019; Teixeira et al., 2019)
Ground granulated blast furnace slag	Cement	(Das et al., 2021; Gholampour et al., 2021; Kim et al., 2021; Nanayakkara et al., 2021; Al-Yaqout et al., 2020)
Metakaolin	Cement	(Caldas et al., 2021; Xie et al., 2020; Bassani et al., 2019; Vejmelkova et al., 2018)
Rice husk ash	Cement/fine aggregate	(Jittin et al., 2021; Al-Mansour et al., 2019; Billong et al., 2018)
Palm oil fuel ash	Cement	Hamada et al. (2018)
Pumice powder	Cement	Wongsa et al. (2018)
Waste glass powder	Cement/fine aggregates	(Kim and Yang, 2021; Abdelli et al., 2020; Ali et al., 2020)
Brick dust	Cement/aggregates	(Fort and Cerny, 2020; Wong et al., 2018; Navratilova and Rovnanikova, 2016)
Sugarcane bagasse ash	Cement/aggregates	(He et al., 2020; Murugesan, Vidjeapriya, Bahurudeen)
Marble dust	Cement/aggregates	(Aydin and Arel, 2019; Evram et al., 2020; Prosek et al., 2020)
Seashell waste	Cement/aggregates	(Mo et al., 2018; Eziefula et al., 2018)
Waste rubber	Aggregates	(Li et al., 2019b; Strukar et al., 2019)
Construction and demolition waste	Aggregates	(Vo et al., 2021; Robayo-Salazar et al., 2020)
Mine waste	Aggregates	(Gallala et al., 2021; Gao et al., 2020; Zhang et al., 2020a)
Plastic	Aggregates	(Almeshal et al., 2020; FauzanNur et al., 2021; Akinyele et al., 2020; Basha et al., 2020)
Biomass ash	Cement/aggregates	(Ohenoja et al., 2020; Rissanen et al., 2020; Sakir et al., 2020; Tosti et al., 2020)

3.2.1. Building's operation

The improvements in indoor thermal comfort can be viewed as one of the main driving factors which stimulate stakeholders to application of an additional layer of thermal insulation for facing changing weather conditions. Together with the development of urbanization, the population boom, and increased requirements for indoor thermal comfort, the range of the thermal insulation materials was significantly improved, and the current offer of the insulation materials is broad (Ricciu et al., 2018). Moreover, the efforts dedicated to the improvement of the thermal performance of buildings were also reflected by materials engineers who consider thermal conductivity as one of the crucial parameters for the design of advanced construction materials. In the context of the EPBD targets, the design and development of an efficient thermal insulation system is the primary goal for materials engineers and architects (Kalaycioglu and Yilmaz, 2017). Hand in hand with this goal comes a detailed understanding of the subsequent goals such as detailed knowledge of envelope inertia (Ricciu et al., 2018), material parameters (Pavlik et al., 2015), understanding of the relationship between hygric and thermal parameters (Cai et al., 2017), measuring techniques (Bertone et al., 2018; AlFaris et al., 2016; Conticelli et al., 2017), and material applicability (Walker and Pavia, 2015).

The application of insulation materials can be viewed as the most efficient way towards a decrease of energy consumption and improvement of building's thermal performance (Pacheco-Torgal, 2014). The extensive studies aimed at a variety of thermal insulation materials (Jelle, 2011; Abu-Jdayil et al., 2019; Kumar et al., 2020) described the main options for the building practice, such as mineral wool, expanded polystyrene, extruded polystyrene, and polyurethane, together with more advanced materials, e.g., vacuum insulation panels, gas insulation panels, or aerogel insulations. The extensive studies dealing with conventional insulation materials also provided their various features, such as thermal conductivity, mechanical parameters, fire resistance, service life, freeze resistance, water transport parameters, and environmental impact (Kumar et al., 2020; Jiang et al., 2020; Colinart et al., 2020; Perez-Bella et al., 2020). Among the utilization of conventional insulation materials, recent studies also pointed at the beneficial effect of green roof insulation or facades based on numerous evergreen plants which are able to moderate interior temperature as well as relative humidity (Fan and Xia, 2018; Khabaz, 2018; Scharf and Zluwa, 2017; Vox et al., 2018). Another promising technology for the minimization of energy consumption is based on thermal storage systems, particularly the application of phase change materials (Navarro et al., 2019; Memon, 2014). The ability of these materials to absorb and consequently release the thermal energy can avoid extensive heating during temperature peaks without additional A/C devices and maintain the indoor temperature in the desired range. The massive investigations of phase change materials provided favorable results related to the possible energy savings achieved by means of free cooling/heating of the buildings. They can be used as microcapsules that can be easily incorporated into conventional building materials or as a part of the advanced solutions for mitigation of energy consumption (Potential of microencapsu, 2020).

Besides the use of materials leading to the decrease in energy consumption, the development and consequent adoption of thermal performance calculation methods represent another substantial task for overcoming the technological barriers (Meijer et al., 2019). A proper understanding of the thermal behavior of building facades is an essential goal for the identification of the most favorable solution towards improved energy performance of building envelopes (Gaspar et al., 2018). Energy efficiency measures can be divided into several categories, such as passive energy measures, reduction of HVAC energy, usage of renewable energy, and energy management (Tian et al., 2018). The attention paid to thermal energy optimization led to significant research developments aimed at optimization techniques for building design (Elkhapery et al., 2021; Serrano-Jimenez et al., 2021). Many advanced user-friendly tools appeared which can be employed for the definition of new perspectives that are not reachable by using traditional techniques (Machairas et al., 2014). Across the literature, it is possible to distinguish many approaches and a plethora of models that were derived by various authors to provide coherent outcomes (Hashempour et al., 2020; Koci et al., 2019). The recent advances in the development of whole building simulation models are able to help designers, architects, policymakers, and stakeholders in finding an energy-efficient solution based on the existing technologies (Serrano-Jimenez et al., 2021; Pasoni et al., 2021; Chen et al., 2020). These strategies may be of importance since the increasing global temperature will result in changes in heating/cooling patterns according to the geographic area. As investigated by Kočí et al. (Koci et al., 2019), the warming trend in mild climate could change the heating/cooling costs by about 15% in a relatively short period. Considering more pronounced peak temperatures in Mediterranean or Nordic countries, this trend needs to be taken into account in the prediction of future building energy demands within the design stage (de Masi et al., 2021).

3.3. Social acceptance

The climate-change associated issues pose a challenging task also for the current society, since disruption of societal patterns is probable to occur. Therefore, the acceptance and application of sustainability measures present an optimal way for damage prevention. However, resolving such a problematic issue cannot be based on technical disciplines only; it requires a complex and multidisciplinary approach (Warnsler and Brink, 2018). A significant part of the world's population lives in highly urbanized areas and faces changes in the paradigm in urban development and societal patterns. Despite the threats appearing as a consequence of the increase of climatic hazards, the inclusion of social sciences has been mostly ignored and neglected (Araos et al., 2016). Furthermore, no coherent strategy useful for a successful solution to this problem from the social point of view is available. Across the literature, it is possible to identify only minor contributions regarding the potential of social sciences in this field (Haarstad et al., 2018). The merging of the International Council for Science and the International Science Council pointed to the necessity of the interdisciplinary approach for solving and framing problems associated with sustainability transition. The communities' perception and acceptance of the sustainability transition can be considered as one of the most important factors in this process and according to various studies, it requires a cultural shift (Rentschler and Bazilian, 2017). Underestimation of comprehensive consultation and communication, accepted reforms, and procurements can face significant obstructions in the public sector (Bazilian and Onyeji, 2012).

3.3.1. Role of social background

Haarstad et al. (2018) assessed the roles of social scientists in climate and energy transformation and formulated three engagement subcategories: (1) production of relevant knowledge, generation of change-catalyzing insights applicable for sustainability transformation context; (2) revision of the new solutions, enlargement of possibilities framework and identification of novel approaches; (3) identification and creation connection between policymakers, stakeholders, government, social factors, and fragmented processes. Mendizabal et al. (2018) analyzed several approaches contemplating stimulation impulses for transition and transformation of urban areas. The transition of the community can then contribute to the modification of standard urbanization tools to meet sustainability criteria regarding sustainable living, clean energy, and resilience (Schlor et al., 2018; Privitera et al., 2018).

Using advanced methods of analysis of socio-technical systems, the mutual evolution and adaptation of a societal and technical system over time in nonlinear conditions can be described (Huang et al., 2018; Zischg et al., 2019). The description of structural changes in society and its governance can be called transition management, which merges real experiments and complex assessments. The behavior of such a system can be analyzed from the point of view of time-horizon importance and validity, i.e., strategy, tactics, operations, and reflection aspects (Rijke et al., 2013; Nevens et al., 2013). These kinds of systems are usually employed in urban specific areas, such as energy and water distribution, with specific and well-developed principles and guidelines (Yuan et al., 2012). On the other hand, a socio-ecological system deals with an interaction between social and ecological systems with a focus on long-term perspectives with anticipated disruptive changes and the following the recovery processes. The decomposition of such a system leads to the identification of its particular phases. This process can be described in detail as the adaptation cycle. The impulses for changes can be divided into several categories: natural, socio-ecological, or institutional (Sharpton et al., 2020; Huang et al., 2018). The comparison of adaptive and transition approaches performed by Garmestani and Benson (2013) resulted in the promotion of adaptive management (originating from the socio-ecological system) due to its more advanced analytical methodology with a shaper process orientation to sustainability as compared with the transition management.

Warnsler and Brink (2018) analyzed new options applicable in social science practice that can define new pathways towards advances in sustainability transitions. They concluded that new findings from neuroscience and neuroplasticity, psychiatry, psychology, ecopsychology, and education allow the generation of a new variety of options for accelerating the sustainability transition by mindfulness. The definition of this term can be found in various sources (Powietrzynska et al., 2015; Bernal et al., 2018; Fischer et al., 2017), but in general, it is understood as intentional, non-judgemental attentiveness to the present moment (Biesbroek et al., 2014). Based on the results of recent research, the utilization of these scientific disciplines can cause a reconsideration of the way how we think and act at facing a crisis. The studies of Ericson et al. (Wamsler et al., 2018; Ericson et al., 2014) pointed at the usefulness of this approach for revealing a new perspective for the mitigation of climate change. However, follow-up studies need to be performed to provide a qualitative and quantitative evaluation of mindfulness contribution. Thus, the creation of assessment tools for a particular context (geographical, cultural, etc.) can bring more rapid acceptance of changes associated with societal changes as suggested in (Warnsler and Brink, 2018; Rentschler and Bazilian, 2017; Mendizabal et al., 2018).

3.3.2. Role of stakeholders

Although the efforts aiming at the improvement of thermal performance of buildings were considered in many countries during the last decades, the necessary positive attitude of stakeholders was mostly not taken into account in a sufficient way. Moreover, the expenditures were not reflected in decreased energy demands. A positive example can be found in a different field, the clothing industry (Jacobs et al., 2018). Here, the value-attitude-behavior model was used for the identification of the gaps and further improved by psychographic constructs. Notably, price sensitivity was revealed as a barrier to the adoption of sustainable preferences. The object of the study can be hardly compared to the decision-making processes in the building industry. However, this study together with many others points to the development of a comprehensive educational program heading to sustainable development that can improve the positive attitude of the society. The data obtained by Heidrich et al. (2016) revealed a huge underestimation and a lack of proper information in dealing with the adaptation strategies. Mendizabal et al. (2018) identified the relevant socio-economic triggers needed for the achievement of the sustainable vision based on the relevant theories, frameworks, and methods applicable to address climate change in urbanized areas.

The European Union as the world leader in global climate responsibility also thanks to the highly urbanized countries and the huge possible impact of climate changes, created comprehensive National Adaption Strategies (NAS) to facilitate and coordinate these efforts on the national and regional level (Biesbroek et al., 2010). Essential for the formulation of NAS is the understanding of the role of motivators, the factors which are required for motivation and influencing the success rate, and facilitating factors or levers needed for increasing the success rate of the individual adaptation actions. The extreme climate events with a combination of EU policies, together with convincing experiences from other countries, create a demand for the formulation of desired adaption strategies. Tompkins and Amundsen (2008) divided these triggers into two groups: climatic and non-climatic to designate direct and indirect drivers and levers for each particular category. The correct characterization of initial motives in cooperation with the scientific community generates a significant increase of knowledge regarding the key facilitating factors without which it is not possible to perform an adequate reaction. These factors include political will, human resources, financial resources, process coordination between individual elements, key actors, different sectors, mutual compatibility, and sufficient knowledge (Walker et al., 2015). Many studies addressed the description of sustainability social barriers, limitations, and ways how to overcome them (Ford and King, 2015; Adger et al., 2009; Aguiar et al., 2018). These barriers often consisted of poor leadership with limited inspiration skills and reduced long-term political commitments (Fuhr et al., 2018). The poor communication and lack of information then resulted in a perception of climate change as a problem for citizens but not as a task that requires immediate action (Aguiar et al., 2018; Vogel and Henstra, 2015).

3.4. Economic evaluation

The renovation of the current building stock, as well as the building of new energy-efficient houses, is inevitably interconnected with increased costs for stakeholders. This issue poses an eminent factor for the retrofit of outdated houses (Bertone et al., 2018). As described above, the acceptance of sustainable principles needs to be connected with all sustainability pillars to achieve the intended targets. However, despite various national initiatives aimed at speeding up the renovation of outdated and energy inefficient buildings, the success of these strategies is very limited and does not achieve the expected results (Filippidou et al., 2017). Specifically, the EU statistics show that the annual rate of retrofitting ranges from 1 to 1.5% (Hashempour et al., 2020; Moran et al., 2020). However, according to the Energy Efficiency Directive (2012/27/EU), the minimum rate needed for the achievement of 80% reduction of GHGs emissions requires at least a 3% renovation rate. Taking into account the relatively low rate of building new constructions, the risk of failing to achieve the stated objective became very likely. As described in the literature, the increased costs associated with a transition to low carbon homes together with the poor acceptance of life cycle thinking measures pose another constrain for relevant stakeholders (Passoni et al., 2021; Ingrao et al., 2018). To balance and promote environmentally and economically viable solutions, the motivation through a calculation of life cycle costs can be viewed as desired tool (Milic et al., 2019). In other words, the environmentally viable solution in terms of environmental footprint payback must match with the return of investments (Asdrubali et al., 2019). However, the building practice refers rather to a maximization of the economic benefits over truly sustainable buildings retrofitting (Menassa and Baer, 2014). A number of scientific reports point to the complexity of the cost-effectiveness of buildings renovation (Bandeiras et al., 2020; Elkhapery et al., 2021; Bachmann, 2020). While the application of an additional thermal insulation layer can be viewed as the most crucial, particular importance needs to be paid also to the type of the heating system, the service life of the used materials, and other running costs. To provide comprehensive information on the relationship between investments and energy-savings steps, several approaches can be found in the present literature.

3.4.1. Service life cost analysis

The application of life cycle cost analysis (LCC) is being viewed as the most suitable appraisal method in the building sector that provides reliable outcomes and relevant information for investors (Petit-Boix et al., 2017). This concept reflected in the Directive 2010/31/EU and completed by the discounted cash flow method is a substantial part of the decision-support tools heading to the implementation of energy efficiency measures of buildings (Leskinen et al., 2020; Amstalden et al., 2007). However, only single LCC optimization often refers to diverse results as described in the study of Milic et al. (2019), who pointed out the limited savings in the case of deep energy renovation. This conclusion complies with the findings that although 30% of the dwellings were renovated, only a minor part had major energy improvements (Filippidou et al., 2017).

Despite the advances in LCC analysis during the last decade, it still struggles with significant uncertainty in parameters, such as discount rate, energy price expectation, and inflation rate (Zheng et al., 2019; Schmidt and Crawford, 2018). Compared to other fields where LCC is widely applied, the assessment of buildings requires consideration of the mentioned variables in the long-term horizon, usually in a range between 30 and 150 years (Rauf and Crawford, 2015). Naturally, the prediction of LCC indicators, such as net present value (NPV), net present cost, or payback period competes with a significant uncertainty (Christensen et al., 2018). On the other hand, some doubts about adopting NPV as a universal evaluation tool for environmental projects refer to the overestimation of conservation costs. In fact, accepting the NPV as a universal indicator may inflict overestimation of optimal solution based on welfare priority and hinder environmental and social criteria (Knoke et al., 2020). As outlined by Cabo et al. (2020), the main disparity lays in the question if the consumption of natural resources to maximize economic growth is acceptable when facing long-term consequences of climate damages and thus limited future GDP. As broadly discussed within the Stern review (Liu et al., 2021b; Nordhaus, 2007; Weitzman, 2007) and following reactions, there is no overall agreement on discounting the long-term project despite its particular importance in facing climate change. Opposed to a single value of discount rate, non-constant discounting was introduced in order to provide a framework for environmental protection policies (Cabo et al., 2020; Gollier, 2010). However, such discussion can be found in economic journals only and is very limited in other areas, such as the construction industry.

3.4.2. *Long-term discounting rate*

The available literature on building evaluation shows that the issue of discounting is not addressed at the ethical level (Hellweg et al., 2003) and mostly consists only in choosing a certain rate in connection with other similar studies, localities, or other investment projects. Long-term forecasts lay beyond the field of traditional techniques used as decision-supporting tools (Merli et al., 2018; Santangelo et al., 2018) and are distorted by substantial uncertainties. In order to provide more reliable values of used LCC indicators, the adoption of a suitable discount rate represents a rule of the thumb. However, the used discount rate in published studies varies between 0 and 15% according to the location and type of the building (Milic et al., 2019; Toosi et al., 2020; Zhang et al., 2020b; Islam et al., 2015). Considering the summary published by (Copiello et al., 2017), the limited framework of appropriate selection of the discounting rate further increases the uncertainty of performed analyses and increases the investment risk. On top of that, the discount rate was found as more influential over the energy price and the energy inflation rate (Zheng et al., 2019). Nevertheless, despite its particular importance, there is no overall agreement on which discount rate should be applied. As remarked by Toosi et al. (2020), even discounting by a rate higher than 8% can be found in the literature despite the conclusions of de Vasconcelos et al. (de Vasconcelos et al., 2016), who investigated the selection of discount rate, and found the rates higher than 6% as prohibitive for policies aiming at improvements in energy performance. On the other hand, discount rates as low as 0.37% were applied in Slovenia for a 10-year run (Kusar et al., 2013).

The uncertainty projections in the long-term run pose an extremely challenging task even in the case of employment of combined expert forecasts and econometric approach (Gillingham et al., 2018). A partial improvement can be achieved by implementing the fuzzy-based approach to decrease the level of uncertainty and include a variety of other parameters, e.g., performance degradation, the residual value of building parts, or policy changes (Ruparathna et al., 2017). Notwithstanding, substantial attention should be paid to discounting due to its prominent role over other time-dependent variables. As concluded by Copiello et al. (2017), the discounting rate is about four times more influential compared to the energy price and energy inflation rate.

3.4.3. *Monetization of environmental externalities*

A very efficient tool for decision support that can bridge the ambiguity between environmental and economic factors can be viewed in the monetary evaluation of calculated environmental indicators (Pizzol et al., 2015). In this regard, several relevant monetization methods can be distinguished in current literature, such as Environmental Priority Strategies (EPS), LIME3, Environmental-Costs/Value Ratio (EVR), Ecotax, MMG, Trucost, Ecovalue12, and Stepwise2006, which were developed to define the magnitude of environmental burden. Undoubtedly, the coherence of all methods is questionable, so boundary conditions, geographical reference area, and other variables need to be defined properly to avoid misrepresentation of obtained results. As concluded by Arendt et al. (2020), the evaluation of particular indicators differs substantially. While the evaluation of human health impact is well developed and shows coherent results across the used monetization methods, the biodiversity indicator calls for clarification. However, only a limited number of research studies were aimed at understanding biodiversity evaluation (McCarthy et al., 2012). A similar effort was in past focused on the definition of social costs of carbon dioxide emissions but this concept does not cover all environmental issues associated with human activities (Bachmann, 2020; Boyce, 2018). On the other hand, the obtained knowledge in the sense of a methodology approach can be at least partially utilized for other impact categories (Tol, 2018; Nordhaus, 2017).

4. Results and discussion

Climate change and its consequences are accompanied by turbulent changes that will probably affect everyone in the upcoming decades. Therefore, they require not only a sufficient level of expert knowledge but also an understanding of efficiency principles by all participants and adequate financial support.

Almost all European countries applied various instruments aimed at the promotion of dwellings renovation. Notwithstanding, the renovation rate still did not reach the desired level (Li et al., 2019a) and remained on the level of natural innovation driven by the service life of buildings. France and Denmark show the best results in this context but only 8% and 7%, respectively, are labeled by A energy performance category while the major share of the building stock still remains in D or even worse categories (Fig. 4). In other words, most of the current building stock exhibits more than 208 kWh/m^2/year primary energy consumption (European commission.B, 2014), in defiance of campaigns, subsidies, and other promotion tools. Apparent differences in the listed values are caused by the climate, housing type, and occupants' requirements on thermal comfort.

4.1. *Barriers characterization*

Fig. 5 summarizes the major barriers to the adoption of sustainable principles in the building industry that prevent rapid renovation of the current outdated building stock in the EU. The collected data consider their importance and knowledge available in the literature. The identified barriers are also labeled by color to particular fields (according to the sustainability principles depicted in Fig. 1) and with relevance to a particular stakeholder group. Apparently, the results cannot suggest which barrier is the most important one, since all barriers are closely related, affect different stakeholders, and belong to various fields. In fact, the main point of this study refers to the finding, that the proposed solution of one particular barrier does not provide any significant step towards sustainability in the construction industry. While the availability of advanced materials as well as environmental assessment tools cannot be deemed as the main barriers, the limited acceptance of such measures by relevant stakeholders poses a substantial issue that should be reflected. However, as all identified barriers are interconnected, it is very difficult to call a particular barrier the most crucial, because it is an integral part of several consecutive barriers that cannot be addressed separately. It is though possible to identify how these barriers interfere in some cases, affect more stakeholder groups while being from different fields at the same time, which reduces their grasp and the possibility of effective solutions. In this sense, the missing link mentioned in the manuscript title consists of the comprehensive approach meeting various research fields, and diversity in approaches of individual groups of stakeholders.

These barriers can still be considered on a more detailed level, according to the country, or type of the building. The suggested solutions, as described, e.g., by Jagarajan et al. (2017), lie in the implementation of sustainability principles by political decisions, availability of expertise

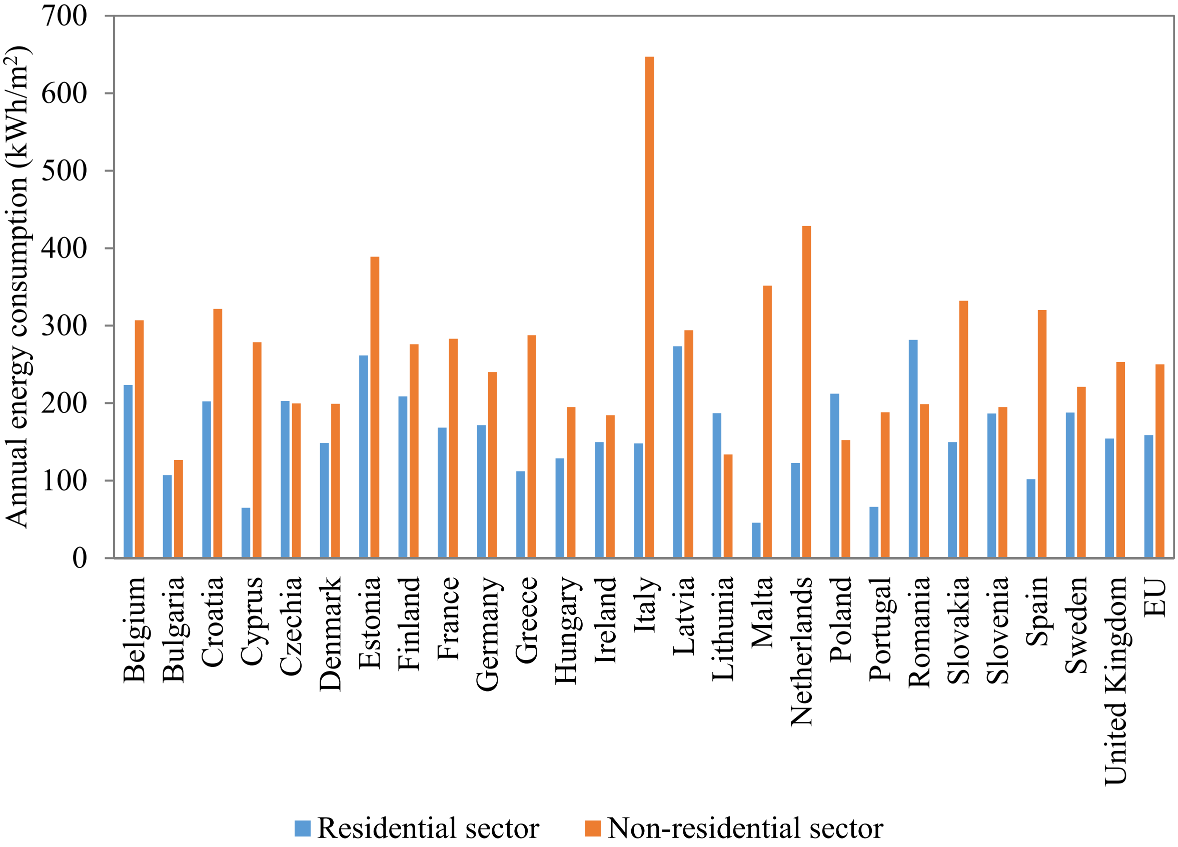

Fig. 4. Average annual energy consumption of residential and non-residential buildings in EU.

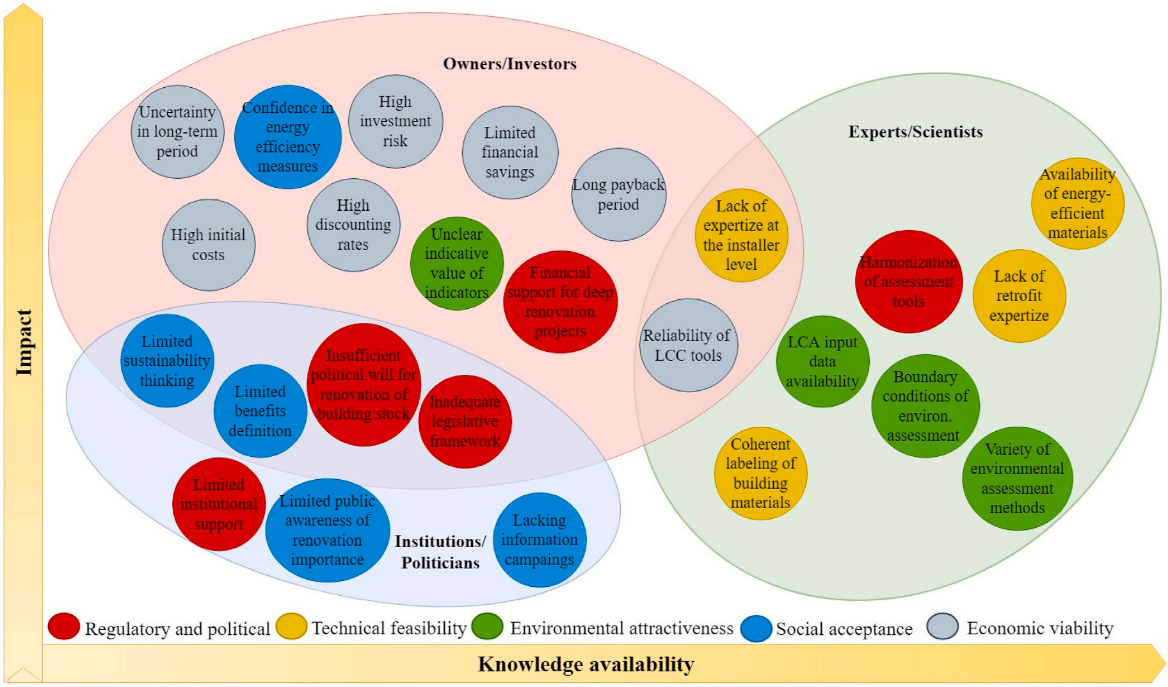

Fig. 5. Impact and relevance of identified barriers.

and relevant knowledge, clear communication to stakeholders, and broader application of cost-benefit analyses. However, the analysis performed in this review reveals that the importance of the interdisciplinary approach remains hidden and is not sufficiently emphasized.

4.2. Multidisciplinary implications

The construction industry has historically always been seen as an engineering field that relies mainly on a precise design of used building elements with a small overlap to areas, such as ergonomics, design, financial viability, or technical feasibility. Facing the advances in buildings design and complex requirements of buildings performance, the change in the paradigm demands integration of various scientific disciplines. The expert knowledge cannot be restricted to the building design only. Experts from the other fields, such as environmental management, logistic, economy, communications, and sociology, need to be included in the preparation of actions plans. These plans can then serve as a knowledge base for politicians which is supposed to be complemented by a legislative framework and support projects that focus not only on funding but also on understanding the importance of the issue. The insufficient interdisciplinary cooperation can be further illustrated by discounting of building renovation projects. While the discussion about the selection of discount rate has been going on for at least two decades, the papers focused on the LCC of reconstruction projects did not adequately reflect this fact despite its undeniable significance. A similar gap in the multidisciplinary approach can be found in lacking public understanding of energy efficiency measures and the particular importance of this issue. Reaching the agreement in several specific questions needs to be supported by the political will to apply sustainability principles to a greater extent by strategic planning taking into account the long-term horizon of these incentives.

Thanks to the relatively well-developed methodological frames of LCA, the process-based LCA is involved in more than 70% of environmental analyses carried out all over the world (Vilches et al., 2017). However, regarding the territorial origin of these methods, datasets and other variables are usually addressed to the Western Europe or USA regions (Pieragostini et al., 2012; Alberti et al., 2017). Therefore, the reliability of the results from other regions is significantly reduced. The quality of the LCI data (originated in the above-mentioned regions) is highly important for produced outputs and a significant effort has to be devoted to developing LCI data for other regions, such as India, China, Australia, Africa, or the Middle East, to improve the accuracy and validity of performed studies (Hossain and Poon, 2018; Edelen et al., 2018). Notwithstanding, substantial advances in materials science, as well as building design tools, allow substantial improvements in the thermal energy performance of buildings and customization of the used materials with emphasis on less-energy intensive materials.

Contrary to the advanced environmental assessment tools with a consistent methodology, the life cycle costing still poses a challenging task. On top of that, credible outputs are based on a variable that cannot be easily adopted. A major drawback, compared to the environmental assessment, lies in a very limited interdisciplinary cooperation. As reflected in the economic journals, the question of ethical discounting, or prescriptive/descriptive discounting has been discussed for at least 20 years, with Stern's review making the subject an almost explosive topic. Moreover, the discussion about the non-constant rate in case of long-term implications took place as well. However, these trends are reflected very poorly in manuscripts dealing with the building renovation, optimization of long-term performance, prolongation of service life, etc. The single value in a very broad sense is used rarely and, the worst, a very limited discussion about the above-mentioned issues can be found in available manuscripts in the civil engineering or construction and building technology journals. Taking into account the very low thermal performance of buildings and unsatisfactory renovation rate, the fulfillment of ambitious goals will be extremely difficult to achieve because, without a truly multidisciplinary approach to this issue, there will be no significant advance in overcoming current barriers. Given the current circumstances and knowledge, substantial progress can be attained by finding the agreement on crucial aspects supported by interdisciplinary cooperation as a decision-supporting tool for government policies. Including intergeneration effects on current actions in terms of environmental policy, the equilibrium between both the generation benefits and project costs should be defined (Liu et al., 2021b). Great importance lies in the harmonization of methodologies and procedures, underpinned by political commitments and will to provide a coherent approach to this important issue.

The social pillar of sustainability can integrate various challenges that were mostly neglected in the past. The increased attention paid to the understanding of global climate changes and following actions revealed a lack of knowledge in the sense of transformation and adaption process. The recent studies showed though some prerequisites for overcoming those barriers, which are mostly related to insufficient political leadership reflected in incoherent regulatory frameworks, information barriers between involved participants regarding the urgency of acting, and underestimation of the role of social sciences.

4.3. Recommendations for future research

The findings gathered in this review paper, together with the identified gaps in the literature and the implications for sustainable-meeting strategies, showed that the transition towards a more sustainable building sector can be boosted by addressing properly several principal issues in future research:

- A comprehensive and multidisciplinary approach based on the consensus in the essential aspects of the individual areas (social acceptance, economic viability, natural environment, technical feasibility) should be created and effectively applied for the definition of a new objective function as the complex assessment tool covering all relevant variables.
- The development of quantified social measures for sustainable development in the building industry presents an actual problem, which needs to be studied in detail. Otherwise, these measures cannot be used effectively as input parameters for advanced decision-support tools.
- As the limited public acceptance of the current incentives can be considered as a limiting factor for the necessary political support, the proper identification of relevant stakeholders and finding appropriate methods of communication presents another important problem to be solved. In that respect, the building sector transition should be seen also as a socio-psychological problem viewing technical and environmental arguments as a knowledge and support base.

5. Conclusions

The primary motivation of this article was to emphasize the complexity of sustainable building retrofits in the residential sector. Although there is a number of tools in the individual disciplines, which are not sufficiently utilized or sufficiently known in other fields, the decomposition of the whole process into individual subproblems is not a strategy that leads to the desired effect. The process in its entirety represents a challenging task, an integral part of which is the change of the technical paradigm to a multidisciplinary approach involving a number of factors, which were underestimated to date, particularly social issues, ethical (intergeneration context), or their acceptance among stakeholders.

Sustainable development, viewed as the long-term commitment to society, natural environment, and economy, is one of the most often used terms in scientific manuscripts. However, too narrow research boundaries associated with the researchers' specializations did not allow yet to achieve the desired outcomes in the sense of breakthrough findings. The position of the construction sector in terms of energy intensity and consumption of natural materials did not change significantly, despite the enormous efforts devoted to these challenges during the last decades and all the pursuits aimed at the improvement of buildings thermal performance, replacement of natural materials, and employment of advanced processing technologies. These strategies will be of particular importance since the increasing global temperature will result in changes in heating/cooling patterns according to the geographic area. The experience based on the identification of major barriers shows that the searching for adequate solutions to the problem using its decomposition may be viewed as a misapprehension of the overall complexity and the interconnectedness of various factors.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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4 Conclusions and directions of future research

The list of published papers refers to the identification of principal issues related to the "greening" of the construction industry together with proposed research lines that provide new perspectives in this field. Rationalization of the material flows, identification and characterization of suitable waste materials, and development of new building materials represent important scientific fields that became dominant facing current issues with shortage of raw materials, and high prices of energy-intensive binders. In the light of mentioned facts, a new paradigm related to the utilization of natural resources, waste-, and by-products needs to be adopted on a broader scale. Besides the development and detailed characterization of novel materials, theoretical aspects associated with the sustainable assessment of modern materials are formulated as well. The concepts based on combined indicators provide a more advanced assessment and lead to a better understanding of the interconnection between particular sustainable pillars.

The main contributions can be summarized as follows:

- The obtained knowledge contributes to the reduction of the dependence on natural resources and mitigation of the environmental footprint related to cement- and geopolymer composites' production. Moreover, the revealed findings promote the acceptance of selected waste materials as future resource banks.
- Novel approaches should be accompanied by a robust description and environmental labeling of materials by advanced combined indicators reflecting the aspects from multiple disciplines to avoid one-sided solutions.
- The current barriers in the building industry require cooperation between various scientific fields to reduce costs and boost the social acceptance of new materials.

Based on the gathered knowledge, the following main directions of future research can be formulated:

- In the field of efficient use and recycling of material resources the identification and experimental analysis of available waste material sources, with emphasis on construction and demolition waste (precursor) and chemical industry (alkalis) to secure closed-loop recycling is necessary. Considering the recent advantages in digitalization of the building stock, the information regarding the end-of-life buildings and the materials composition can be effectively utilized for the production of new building materials through on-site selective demolition. In these terms, the current building stock can be accepted as an important future resource bank. Such an approach will result in a significant reduction of produced waste, efficient management of material resources in the sense of maximum recycling, and preservation of natural resources.
- In the field of the development of integrated assessment of building materials in terms of sustainable development, it is necessary to accept that the prevailing paradigm based on the single-angle approach does not provide sufficient advances to overcome current barriers. On this account, the available assessment tools should be improved and integrated with recent knowledge advances in the fields, such as economy, environmental protection, and social sciences to meet the ambitious sustainable targets. These goals require a broad interdisciplinary approach and the cooperation of scientists from various fields and countries to address and resolve major barriers.

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