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Lightweight alkali-activated materials and ordinary Portland cement composites using recycled polyvinyl chloride and waste glass aggregates to fully replace natural sand

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A R T I C L E I N F O

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A B S T R A C T

Polyvinyl chloride plastic (PVC) and glass waste have proven to be significant environmental concerns considering their restricted reuse and complicated recycling procedures. Glass and PVC waste materials form a substantial portion of total solid wastes that negatively influence the environment. This study aims to fully replace natural sand with recycled PVC and waste glass aggregates in alkali-activated materials (AAMs). A comprehensive testing programme was employed to investigate the effect of 100 % aggregate replacement on the composites' mechanical performance, water absorption, impact resistance, thermal conductivity, resistance to harsh environments, and microstructural changes. Results revealed that AAMs containing recycled PVC and glass aggregates outperformed their ordinary Portland cement (OPC)-based composite counterparts in terms of mechanical properties, energy absorption, thermal conductivity, and carbon footprint estimation. Although mixtures containing recycled aggregates cannot be deemed for load-bearing applications, these composites exhibited a promising capacity to be used in insulating applications. AAMs containing 100 vol-% PVC aggregates with flexural and compressive strengths of 9 and 11 MPa, respectively, registered the highest energy absorption of about 6 J, three times higher than the AAM control sample, and the lowest thermal conductivity of about 0.5 W/mK, with about 80 % reduction of thermal conductivity compared to the AAM control sample. With the full replacement of PVC and glass aggregates, the most significant decrease in the carbon footprint is achieved for AAM (−352.25 kg CO2-eq) and OPC (−353.94 kg CO2-eq), respectively.

1. Introduction

Natural aggregates, gravels, and river sand are essential for construction. Excessive natural aggregate extraction has detrimental physical and hydraulic effects on the ecosystem [1]. The annual global consumption of aggregate used in concrete is between 26 and 30 billion tonnes, with sand and gravel accounting for 65 % to 85 % of the total consumption [2]. Concrete is the most widely used material in the construction industry, accounting for around 25 gigatonnes per year and 3.5 tonnes per capita [3]. Due to the high demand for sand from the construction sector, researchers started to look for low-cost and readily available alternatives to replace natural sand. Moreover, in a recent study by the authors, the microstructure of PVC particles revealed a rough, rounded surface with grooves and notches that provide a tight interlock between the aggregate and matrix. Consequently, the mechanical properties' delamination phenomenon was limited in AAMs with PVC aggregates [4].

The integration of glass waste as a partial replacement for sand in concrete mixtures exhibited adequate compressive and flexural strength and elastic modulus levels compared to the control, which refers to the

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pozzolanic action of amorphous silica in the glass.[5]. Moreover, replacing 0–60 % of aggregates in concrete with 4–16 mm waste glass did not significantly affect the workability of concrete but slightly decreased its strength. [6].

Many materials like PVC, waste glass, sawdust, wood chippings, and wood shavings were utilised in manufacturing concrete mixtures, where PVC, in particular, has shown excellent performance in terms of mechanical properties [7]. In 2016, the global production of PVC surpassed 45 Mt, and it is continually rising [8]. Unlike other polymers (such as Polyethylene terephthalate and Polypropylene), PVC is difficult to recycle and could pollute the environment in several ways if not adequately treated [9]. It is one of the largest sources of dioxin, a highly hazardous persistent organic pollutant (POP), which is created due to insufficient combustion of PVC during its manufacturing process [10]. Moreover, PVC content in municipal plastic waste typically ranges between 7 wt-% to 10 wt-% of the total waste [11].

On the other hand, waste glass accounts for more than 5.8 % of the global Municipal Solid Waste (MSW), where over 14 million tonnes of waste glass are generated in the European Union and 11.38 million tonnes are generated in the United States [12]. These figures demonstrate the global availability of waste glass and advocate its usage as a raw resource. Hence, the employment of waste glass in concrete mixtures as a substitute for cement or natural sand could reduce its disposal in landfills. Moreover, reusing waste glass as a construction material is becoming a viable strategy for reducing the dependency on Portland cement (OPC) and sand as sole sources for binder and fine aggregates [13]. Reusing glass as an alternative to concrete constituents would also reduce CO_2 emissions associated with cement production [13].

OPC-based binders are extensively used as a primary construction material across the world due to their low cost, ease of production, and ability to be moulded into various dimensions or forms, as well as their high compressive strength. However, its brittleness in response to strain or deformation, which leads to the production of many microcracks, teamed up with its intrinsic porous microstructure, might lead to the entry of several noxious chemicals [14].

OPC production is the world's third most significant source of CO_2 emissions, contributing to nearly 10 % of global anthropogenic carbon dioxide emissions. The calcination of limestone is primarily responsible for these emissions, as it takes 1700–1800 MJ of energy per tonne of clinker to reach calcination temperature (around 1500 °C) [15]. On a high note, alkali-activated materials have the potential to emit up to 5 to 6 times less CO_2 than Portland Cement [16]. These alkali-activated materials, known as 'geopolymers,' can be made from various aluminosilicate precursors with varying availability, reactivity, cost, and value. However, unlike Portland cement, alkali-activated materials are far from being a one-size-fits-all answer for meeting future construction material demands. Still, this class of materials is dramatically versatile and responsive to local conditions, with high expectations to become a significant component in the future sustainable building materials business [17].

In a recent study by Lenka et al. [18], cement, fine aggregate, and coarse aggregate were replaced simultaneously by ground granulated blast furnace slag (GGBFS) and lime, granulated blast furnace slag (GBFS), and recycled coarse aggregate (RCA), respectively. Concrete mixtures including up to 75 % RCA and 50 % GBFS, as well as 60 % GGBFS and 6 % hydrated lime, were found to have higher compressive strengths than those stipulated for M20-grade concrete. According to durability testing, these mixes exhibited equivalent or greater resistance to sulfuric acid and chloride ion penetration, a favourable cost-benefit ratio, and much less environmental effect than ordinary concrete. Furthermore, the strength and durability of fibre-reinforced self-compacting concrete were investigated in a study conducted by Ortega et al. [19], where Electric Arc Furnace Slag (EAFS) and limestone fines (0/1.18 mm) were used as aggregate and Ground-granulated blast furnace slag (GGBFS) was used as a binder in concrete. The use of metallic or synthetic fibres decreased the durability of the concrete, thereby increasing the entry of aggressive external agents. Despite the increase in porosity, the improved flexibility of the cementitious matrix due to the addition of GGBFS was advantageous against moist/dry and sulphate-attack phenomena. Gao et al. [20] used molybdenum tailings to substitute fine aggregate in structural concrete. They concluded that the examination of economic and environmental effects found that concrete-filled steel tubes are a preferable option to employing molybdenum tailings as aggregates in structural concrete, especially with a higher replacement ratio of molybdenum tailings.

The bond between the aggregate and cementitious matrix might vary depending on the kind of aggregate used. For both limestone and the electric arc furnace slag (EAFS) aggregates, the development in the interfacial transition zones (ITZ) system's elastic stiffness over time was the same; hence the increase in the adhesion depends on the cementitious matrix's composition [21]. The use of hydrated lime increased the compressive strength of recycled aggregate concrete (RAC) composites containing high-volume ground granulated blast furnace slag (HV-GGBFS). This increase in strength is due to the activation of HV-GGBFS in the presence of lime [22]. Moreover, because of the smaller particle size compared to typical cement clinker, GGBS works on reducing the flowability of self-compacting concrete (SCC), resulting in more uneven dragging of aggregate particles [23]. The compressive strength of concrete with high-volume ground granulated blast furnace slag (GGBFS) and recycled coarse aggregate (RAC) composites increases with the addition of up to 7 % lime, which may be regarded as the optimal dose of lime [24].

Researchers have worked effortlessly to reduce CO_2 emissions and mitigate the solid waste problem. In a study by Senhadji et al. [25], scrapped PVC pipes were utilised in OPC-based mixtures as a partial replacement for traditional aggregates, replacing natural sand and coarse aggregates in proportions of 30 %, 50 %, and 70 % by volume. They concluded that as the replacement ratio increases, the workability of the mixture improves. A significant reduction in concrete's mechanical strength was also observed when sand was replaced with 50 % and 70 % PVC. Despite this loss in mechanical strength, the obtained mixtures were found to comply with the recommendations of the *ACI*-213 standard for producing and using lightweight construction materials (class II structural concretes). Finally, they confirmed that using PVC to substitute sand and aggregates significantly reduced chloride ion penetration through concrete. Similar results were reported by Merlo et al. [26], where a maximum reduction of 50 % and 30 % was observed in compressive and flexural strengths, respectively, when replacing sand with 5 % PVC. The difference in characteristics and properties between PVC and natural aggregates might be responsible for the loss in mechanical strength.

On the other hand, Kim et al. [27] investigated the replacement of natural sand with 50 % and 100 % recycled cathode ray tube (CRT) glass containing heavy metals in OPC-based mortar. They found that the strength of mortar mixtures containing CRT dropped as the proportion of waste glass in it increased. Reduced adhesion between the waste glass surface and the cement hydrates may have contributed to the compressive and flexural strength drop. However, CRT particles were observed to enhance the mixture's resistance to freeze-thaw, chloride ion penetration, and sulphate attacks.

The novelty of this study lies in the complete replacement (i.e., 100 %) of natural sand with recycled PVC and waste glass aggregates in alkali-activated materials to develop lightweight building blocks. Incorporating recycled PVC and waste glass aggregates in OPC-based composites has also been investigated for comparison reasons, and they were employed as a benchmark. The complete replacement of natural aggregates with recycled PVC, combined with the utilisation of Portland cement-free alkali-activated materials (AAM), is a radical approach to adopting an environmentally friendly, circular construction concept. To the best of our knowledge, no study has investigated the complete replacement of natural sand with recycled PVC aggregates in AAM.

2. Experimental framework

Fig. 1 illustrates the experimental framework used in this study to comprehensively investigate the impact of recycled aggregates on cementitious composites' performance.

2.1. Materials

The alkali-activated cementitious composite in this study is composed of the following:

(i) Fly ash (Cemex, UK) complies with the normal fineness (N) requirements of the British standard BS EN 450–1:2012.
(ii) Ground granulated blast furnace slag (Hanson Heidelberg Cement, UK) following the EN 15167–1 requirements.
(iii) Quartz silica (Sika).
(iv) Graded sand with particle sizes ranging from 0 to 0.5 mm to 0.5 – 1.0 mm per the BS EN 410–1:2000 guidelines.
(v) 3.23 mass ratio of sodium silicate (Na2SiO3) solution to SiO2/Na2O (Solvay SA, Portugal) Sodium hydroxide (NaOH) solution (10 mol/l) (Fisher Scientific, Germany).
(vi) Nano-clay of attapulgite (supplied by Attagel 350, Lawrence Industries Ltd., UK).
(vii) Portland-limestone Cement type CEM I (Cemex, UK), complying with EN 197–1, was used in this study to produce the OPC mixtures.
(viii) PVC aggregates supplied by AlterEco (Padova, Italy) were used to substitute natural aggregates. They were ground and sieved to a nominal size of 0.5 mm – 3 mm.
(ix) The waste glass was collected locally from the Brunel University campus recycling point, and then ground to a size range of 0.5 mm – 3 mm.
(x) PVA fibres were provided by Jesmonite, UK, with dimensions of 150 μm in diameter and 12 mm in length.

2.2. Mix formulation and material preparations

A total of eight OPC and AAM mixes were produced (see Table 1). The AAM mixes were produced by mixing all the dry components, including the aluminosilicate source materials (FA, GGBS, and silica), attapulgite (AT) nano-clay additive, aggregates (graded natural sand, glass, PVC aggregates) (see Fig. 2), and 3 % PVA fibre additives, for 5 min at 250 rpm using a planetary mixer (Kenwood, Germany).

Natural sand aggregates were replaced with recycled aggregates (i.e., glass and PVC) at 100 vol-%. Based on the author's prior investigation [28], the dosage of attapulgite nano-clay was kept constant at 1 % by the weight of the binder.

Using a magnetic stirrer, the alkali activator solutions (NaOH and Na_2SiO_3) were mixed for 5 min at a constant mass ratio of 1:2. Premixed alkali solutions were progressively added to the dry mixture and mixed for 10 min at 450 rpm to create consistent AAM mixes. A constant solid-to-liquid ratio of 0.4 was set for all AAMs. Following the mixing procedure, AAM fresh mixes were cast using prismatic moulds with dimensions of $160 \times 40 \times 40$ mm^3 and cured in the oven for 24 h at 60 °C, followed by six days of air curing at ambient temperature.

The four OPC mixes were produced by mixing Portland cement, aggregates (natural sand, glass, and PVC aggregates), 3 % PVA fibres and water. The procedure involved mixing the dry components for 5 min at 250 rpm, then adding water (fixed w/c ration of 0.4) and mixing for 10 min at 450 rpm to obtain homogenous OPC mixes. OPC fresh mixes were cast in prismatic moulds with dimensions of $160 \times 40 \times 40$ mm^3 and cured in water for seven days at room temperature. All samples were tested after 7 days of curing since AAM-based composites reach the optimum geopolymerisation stage at this age, and OPC- based composites reach the earliest hydration age. For both OPC and AAM mixtures, the workability of composites complied with the workability requirements of cast mortars specified in the UNE-EN 13395–1 standard [29].

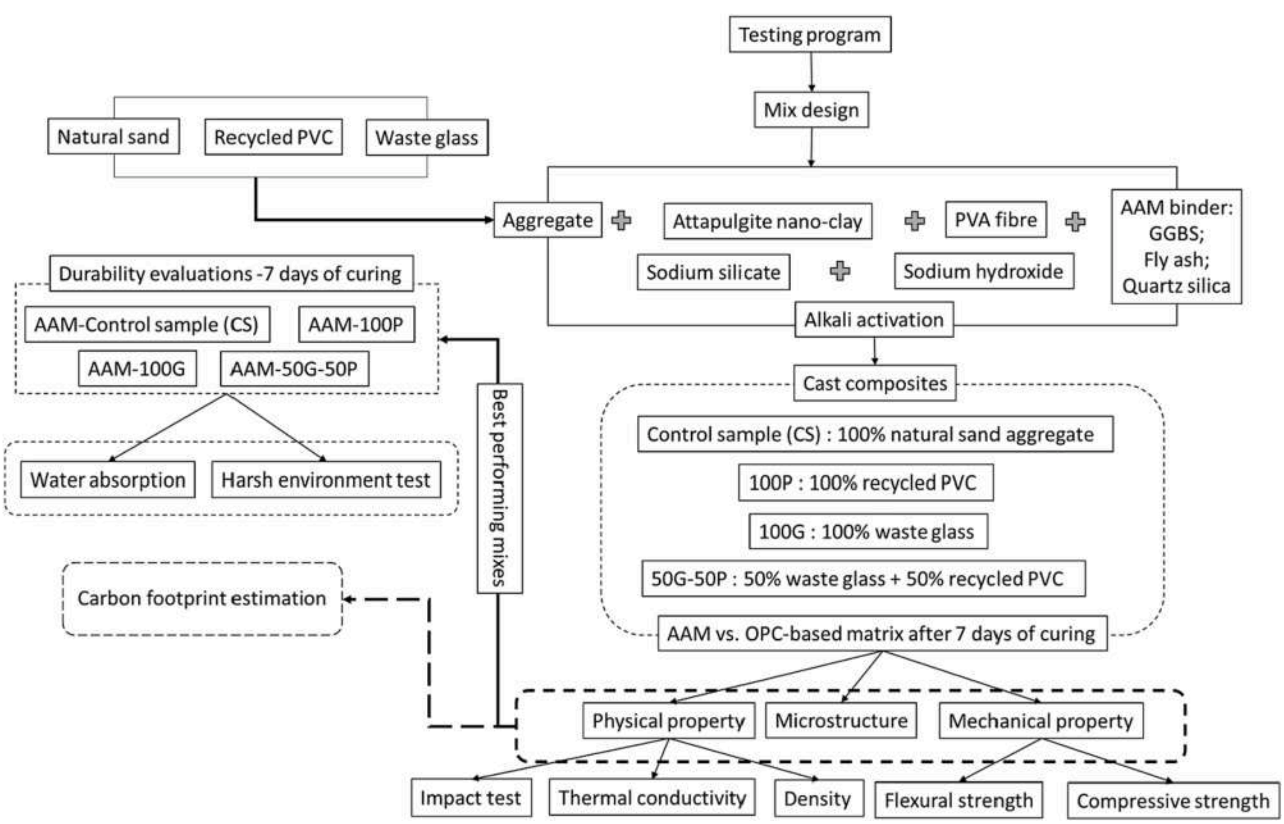

Fig. 1. Experimental design and analysis program.

Table 1
Mix formulations for AAMs and OPC mixtures with PVC and glass at different replacement ratios.

Matrix system	Sample ID	Binder (wt.-%)				Aggregate (Vol.-%)			
		OPC	FA	GGBS	Quartz silica	Natural sand		Recycled aggregate	
						05 – 0.5 mm	0.5 – 1 mm	PVC	glass
AAM composites	CS	0	60	25	15	60	40	0	0
	100P	0	60	25	15	0	0	100	0
	100G	0	60	25	15	0	0	0	100
	50G-50P	0	60	25	15	0	0	50	50
OPC-based composites	CS	100	0	0	0	60	40	0	0
	100P	100	0	0	0	0	0	100	0
	100G	100	0	0	0	0	0	0	100
	50G-50P	100	0	0	0	0	0	50	50

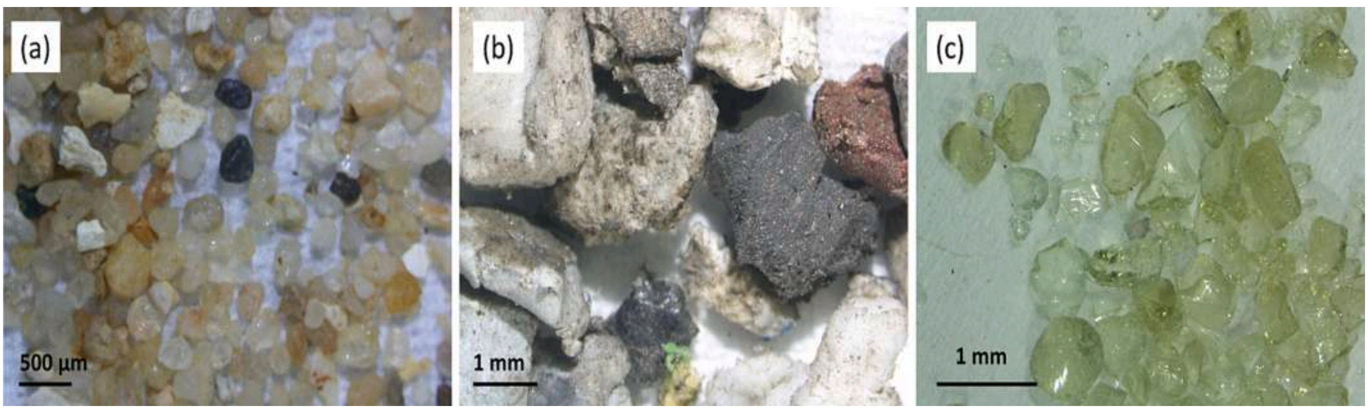

Fig. 2. The visual appearance of (a) natural sand, (b) recycled PVC, and (c) waste glass aggregates.

2.3. Experimental tests

2.3.1. Mechanical properties

The mechanical performance of AAM and OPC-based composites (i.e., flexural and compressive strengths) were tested using an Instron 5960 Series Universal Testing System after seven days of curing, according to the specifications of BS EN 196–1:2016.

2.3.2. Microstructure analysis

The microstructure of AAM and OPC composites was studied using scanning electron microscopy (SEM) (Supra 35VP, Carl Zeiss, Germany). SEM was used to evaluate control samples and AAM and OPC samples containing PVC and glass aggregates. The samples were cut into 8 mm^3 bits and gold-coated using an Edwards S150B sputter coater to improve their electrical conductivity before placing them in the SEM. An average of five samples were investigated for each type of mixture.

2.3.3. Water absorption

The water absorption test was carried out following a modified ASTM C1585-13 procedure. Three prismatic moulds with dimensions of $160\times40\times40$ mm^3 were cast for each mixture composition to serve the purpose of this test. The test was performed on the samples after 7 days of curing. Samples were immersed in water and weighed after 5 min, 10 min, 15 min, 30 min, 1 h, 2, 3, 4, 6, 12, 24, 48, and 72 h from the immersion. The following formula was used to determine the water absorption rate of samples:

$$Water\ absorption(\%) = \frac{M_t - M_0}{M_0} \times 100$$

Where M_0 represents the oven-dried mass and M_t represents the saturated surface-dry mass.

2.3.4. Impact test

Puncture impact tests were conducted at room temperature using an instrumented drop weight tower Instron Ceast 9340 (Instron, Pianezza, Italy) equipped with a CEAST Data Acquisition Systems DAS 64 K. A steel impactor, with a mass of 3.055 kg and hemispherical radius of 12.7 mm, to test OPC and AAM samples. Square specimens ($40\times40\times20$ mm^3) were clamped circumferentially by a steel clamping ring with an inner diameter of 30 mm to prevent movements during impacts. The anti-rebound system of the impact tower was activated to avoid a second undesired impact that could compromise the experimental results, blocking the impactor after the rebound. For each formulation, three samples were tested with an impact energy of 10 J. Results of the impact test were reported in terms of peak energy (E_p), which consists of the energy absorbed via elastic deformation of the specimen and the energy dissipated via damage initiation and propagation [30]. The failure patterns of post-impacted samples were acquired with a digital camera Canon Powershot SX210 IS (Canon Inc., Tokyo, Japan).

2.3.5. Thermal conductivity analysis

A C-Therm TCi thermal analyser (C-Therm Technologies, New Brunswick, Canada) was used to assess the thermal conductivity of the AAM and OPC samples according to ASTM D7984. A current was delivered to the heating element of an alumina sensor, which produced a small quantity of heat. The heat generated causes a temperature increase of 1–3 °C at the sensor-composite contact. The voltage drop of the sensor element changed as the temperature increased at the contact. The thermal conductivity of the composite materials was calculated using the rate of rising in the sensor voltage. Measurements were made at room temperature on $40\times40\times20$ mm^3 samples (three samples per investigated mix), considering two testing points. A contact agent (bidi stilled water) was put between the sensor and the sample to lower the heat resistance to a minimal level. The averaged measurement findings in each sample were used to calculate the thermal conductivity value.

2.3.6. Harsh environment test

AAM samples of 50×50×50 mm^3 were exposed to ten cycles of harsh environmental conditions after 7 days of curing. In each cycle, samples were maintained in a +70 °C water tank for 24 h, then moved into a freezer at −14 °C for 24 h. The weight loss and compressive strength reduction of each cube were then determined.

2.3.7. Carbon footprint estimation

The environmental impact of all mixtures was analysed through Life Cycle Assessment (LCA) in accordance with ISO 14,040 and EN 15804. The analysis is comparative and adopts a cut-off type allocation whereby the environmental impacts of PVC and glass recycling are attributed to the new life cycle. The environmental benefit derived from the recovery of a semi-finished product was subtracted as a waste of a previous life cycle. The SimaPro software, provided by Pré, was used to calculate the environmental impact. The analysis aims to observe the variation of the carbon footprint in the two composites after entirely replacing the sand with PVC or glass aggregates and after replacing the entire volume of sand with 50 % PVC and 50 % glass. The functional unit is a ton of cement mixture, and the system boundaries are defined as "cradle to gate" for each component, thus excluding the environmental impact contributions deriving from their assembly and the cement curing. The geographic allocation of the system is limited to Europe, so the data used represent the average of the processes currently used throughout the European continent. To calculate the carbon footprint, it was necessary to model the production processes of each component. For the representative processes of the production of fly ash, GGBS, silica fume, sand, sodium silicate, sodium hydroxide, OPC, and water, the Ecoinvent 9.3 database [31] was used, which has accumulated datasets related to the single production process in a European allocation. The recycling process of the granulated PVC present in Ecoinvent 9.3 has been suitably modified in a previous work [4] to represent only the recovery, as an avoided product, of PVC produced by suspension (see Fig. 3). Similarly, a process has been defined for the recovery of granulated glass starting from disposed bottles (see Fig. 4), considering as benefit one kilogram of avoided product and as impact the use of 0.0043 m^3 of natural gas necessary for machinery and buildings, in addition to 0.072 MJ of electrical energy used for the grinding operation [32]. Impact values from recent literature were used for the production processes of silica fume [33] and PVA fibres [34] due to the absence of datasets in Ecoinvent 9.3. The environmental impact of nanoclays has been neglected due to the absence of data available for the inventory and the negligible percentage of mass present in the mixtures.

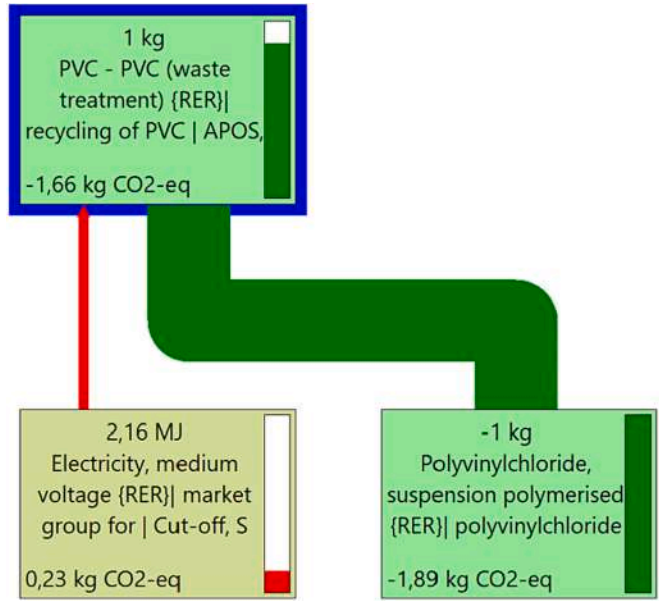

Fig. 4. LCA network of PVC aggregate recycling process.

3. Results and discussions

3.1. Mechanical properties and microstructure

The density and mechanical performance of OPC-based and AAM composites with and without natural aggregate replacements were recorded and compared. One of the critical considerations in this investigation was the density reduction of cementitious composites. It is well acknowledged that the overall weight and, as a result, the density of composites decline when lightweight recycled aggregates are employed as a natural aggregate alternative [35]. Using lightweight cementitious composites has numerous benefits, including reduced production costs, enhanced heat efficiency, lowered environmental impacts, and decreased dead weight of a structure to mitigate the consequences of earthquakes [35]. The results (see Fig. 5a) revealed a substantial reduction in the density of both AAM and OPC-based composites after

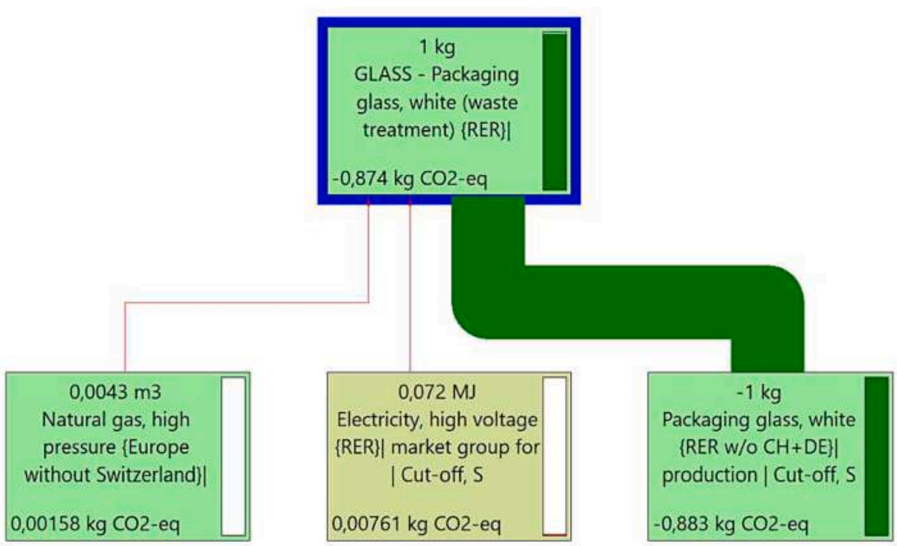

Fig. 3. LCA network of glass aggregate recycling process.

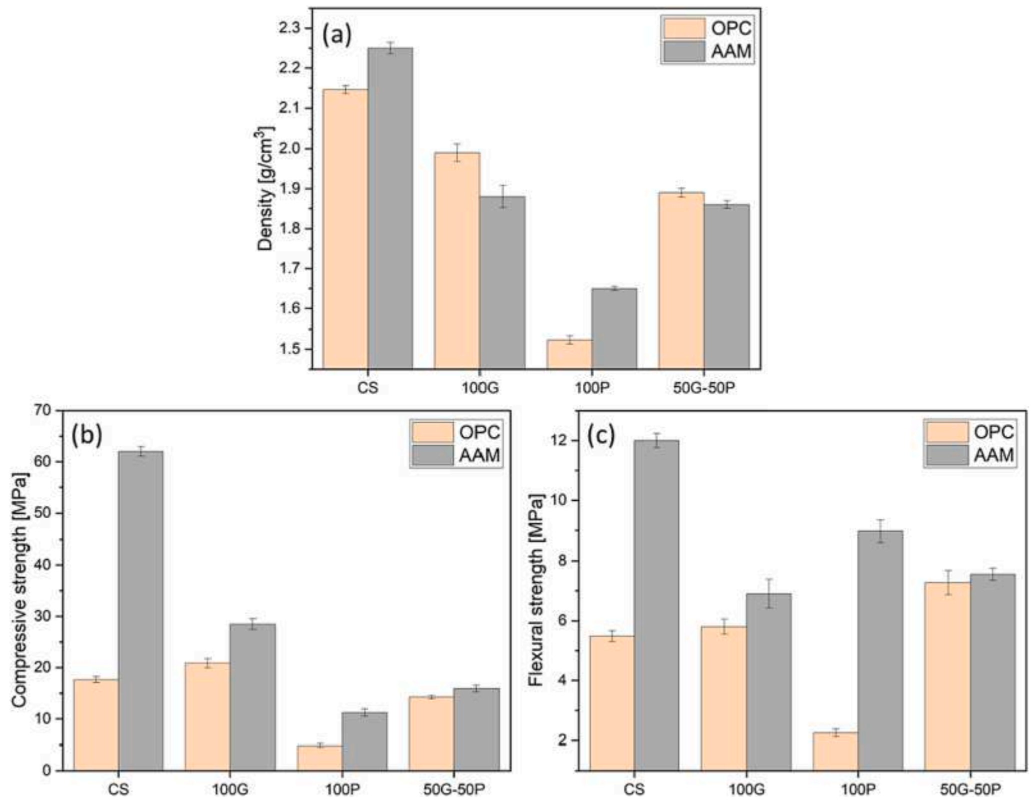

Fig. 5. (a) Density, (b) compressive strength and (c) flexural strength of the cementitious composites.

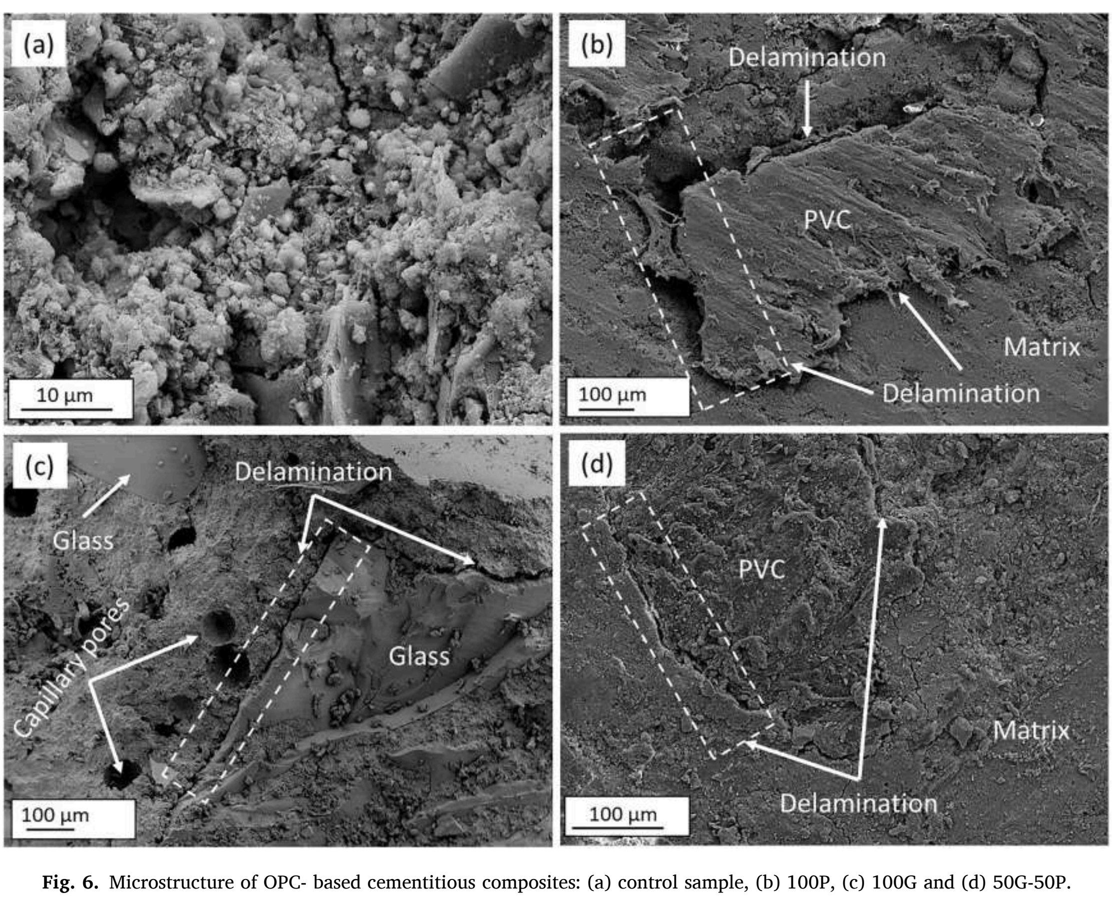

Fig. 6. Microstructure of OPC- based cementitious composites: (a) control sample, (b) 100P, (c) 100G and (d) 50G-50P.

the complete replacement of natural sand with PVC and glass aggregates. As shown in Fig. 5a, maximum density reductions of 27 % and 29 % were registered for the AAM-100P and OPC-100P mixtures, respectively, compared to their corresponding control mixtures. The decline in density is associated with the lower density of recycled aggregates (1.176 g/cm^3 for recycled PVC) compared to the density of the natural aggregate (1.68 g/cm^3), which reduces the unit weight of resulting composites regardless of the replacement ratio and type of substitution [36]. Moreover, the presence of gaps/delamination in the aggregate-matrix interface of composites containing recycled aggregates acts as air voids and reduces the total weight of the final composites [37].

The microstructure of mixtures was evaluated after seven days of curing to detect the development of delamination and the ITZ between the cementitious matrix and aggregates. As evident in Fig. 6 and Fig. 7, apparent delamination, particularly in the composites containing PVC aggregates, was observed, which is developed due to the limited compatibility of recycled aggregates with the cementitious matrix.

The results (Fig. 5b and c) indicated that for both AAM and OPC-based composites, except for OPC-100G and OPC-50G-50P, the substitution of natural aggregates with waste glass or PVC aggregates negatively affected the mechanical performance of samples and considerably reduced the compressive and flexural strength values. Compared to the results reported for OPC-CS, a minor enhancement in mechanical performance (i.e., 18 % in compressive and 5 % in flexural strengths) was observed for the OPC-100G composite. The strength enhancement could be attributed to the microstructural refinement of the composite following the addition of glass aggregates (see Fig. 6a and c).

On the other hand, a remarkable improvement of 32 % was registered for the flexural strength of the OPC-50G-50P composite, whereas its compressive strength was reduced by 19 % compared to the OPC-CS. Moreover, the results also displayed that regardless of the aggregate replacement ratio, AAM composites outperformed OPC-based composites in terms of compressive and flexural strength. It is worth mentioning that the samples containing a combination of glass and PVC aggregates (i.e., AAM-50G-50P and OPC-50G-50P) exhibited negligible differences in both compressive and flexural strengths.

In this regard, the compressive strength of AAM-CS, AAM-100G, AAM-100P, and AAM-50G-50P was about 252 %, 37 %, 131 %, and 12 % higher than their OPC-based counterparts, respectively (see Fig. 5b). As shown in Fig. 5c, a similar superiority trend was also recorded for the flexural strength values, where higher flexural strengths of approximately 119 %, 19 %, 299 %, and 4 % were recorded for AAM-CS, AAM-100G, AAM-100P, and AAM-50G-50P, respectively when compared to the values registered for their OPC-based counterparts. Multiple factors, including the hydrophobic nature of PVC aggregates, the high porosity and air content of PVC composites, the lower elastic modulus of PVC than that of sand aggregates, and the weak interfacial bonding between recycled aggregates and cementitious matrix, hinder the mechanical properties of composites containing recycled plastic aggregates [36].

Previous studies on cementitious composites containing waste glass aggregate have shown contradictory results regarding flexural and compressive strengths. Employing waste glass as fine aggregate with low replacement ratios has been reported to enhance the mechanical properties due to its role in accelerating the hydration or alkali reaction [38]. Contrarily, it has been discovered in various investigations that substituting waste glass for natural sand with high replacement ratios

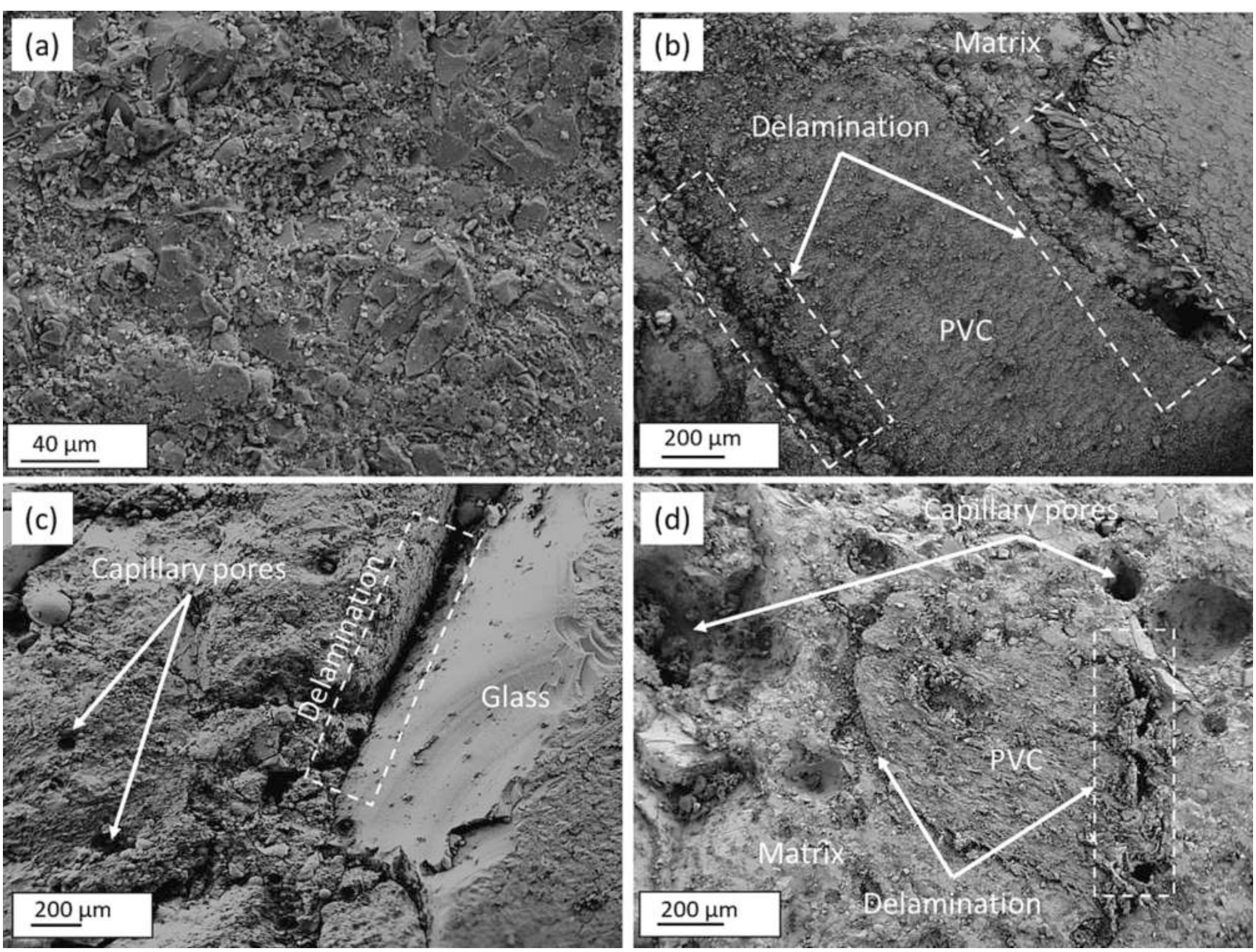

Fig. 7. Microstructure of alkali-activated materials: (a) control sample, (b) 100P, (c) 100G and (d) 50G-50P.

yields a negative impact on mechanical characteristics [39]. The reason is attributed to the surface characteristics of glass aggregate, i.e., the smoother surface of glass aggregates than that of sand aggregates mitigates the interfacial bonding between the matrix and waste glass aggregates [38,40].

3.2. Physical properties

3.2.1. Impact properties and energy absorption

Producing cementitious materials to improve their dynamic and energy-absorbing properties is a much-explored research topic. Despite many advantages, concrete suffers from several drawbacks, such as low tensile strength and low energy absorption capacity, resulting in cracking, spalling, and premature disintegration of the structures. Enhancing the concrete's ductility is a prerequisite for maintaining structural integrity and ensuring the safety and serviceability criteria of the structures [41]. The energy absorption capacity of OPC and AAM composites containing PVC and waste glass as a total replacement of natural aggregates is shown in Fig. 8. A significant increase in the E_p-value was noticed in mixtures when sand was replaced entirely with PVC aggregates. The increments in energy absorption capacity were 31 % and 187 % for OPC-100P and AAM-100P, respectively, compared to respective control samples. The higher elastic properties and flexibility of polymer aggregates than natural ones led to enhanced ductility behaviour in the specimens, leading to increased dynamic energy absorption capacity. Similar experimental evidence was achieved by other researchers, who implemented polymer aggregates to improve the dynamic mechanical properties of concrete [42–44].

Interestingly, the rate of increase found in the two matrices is remarkably different. AAM mixture incorporating 100 % PVC aggregates (AAM-100P) performed better than its OPC counterpart (OPC-100P), reflecting the static mechanical performance reported above. The proper load transfer between matrix and aggregate and non-porous interfacial transition zones (ITZs) are key requirements to ensure an effective contribution of the polymer particles to the dynamic response of the composite. For this reason, PVC aggregates may be better compatible with the AAM binder than the OPC one. Constituents of AAM mixtures, including the alkali activator, could partly assist in enhancing the interfacial bond between the waste plastic particles and the surrounding matrix, as ascertained by the authors in previous work [4]. In support of this assumption, Hu and Xu [45] found that pre-treating recycled PVC particles with NaOH-based solution remarkably improved the mechanical performance of concrete under impact load, as the alkaline solution makes the particles rougher and adds hydrophilic groups on their surface, therefore improving the compaction of ITZ. The ameliorative effect of silica fume in PVC-concrete composites was noticed by Ali et al. [46]. In this work, the authors demonstrated that the pozzolanic and packing effect of quartz silica strengthened the bond between plastic aggregates and the cement matrix. An opposite dynamic behaviour of the two matrices was ascertained by incorporating waste glass in 100 % replacement of the natural mineral aggregate. Compared to CS formulations, AAM-100G showed a 93 % increase in E_p. On the other hand, the influence of glass aggregates on the impact response of the OPC mix was detrimental, bringing a decrement in E_p of about 60 %. In trend with the static mechanical characteristics, this result could be attributed to the different chemical interactions of glass with the two binders. As stated by Si et al. [47], waste glass aggregates would react positively in the geopolymer system, increasing the initial Si/Al ratio, which potentially enhanced the available Si-content for the formation of the reaction products, then resulting in a denser microstructure. On the other hand, it is well recognised that the addition of high glass aggregates content in OPC is strongly limited by the alkali-silica reaction (ASR), which induces stress and severe cracking in the concrete microstructure, worsening its mechanical properties [48]. Hybrid formulations (50G-50P mixes) showed obvious intermediate dynamic behaviour between the 100 % aggregates replaced samples. It is worth highlighting that the partial addition of PVC particles as aggregates in the glass-based OPC mixture (OPC-50G-50P) allows for mitigating the deleterious effect of the waste glass on the dynamic mechanical performance of the composite.

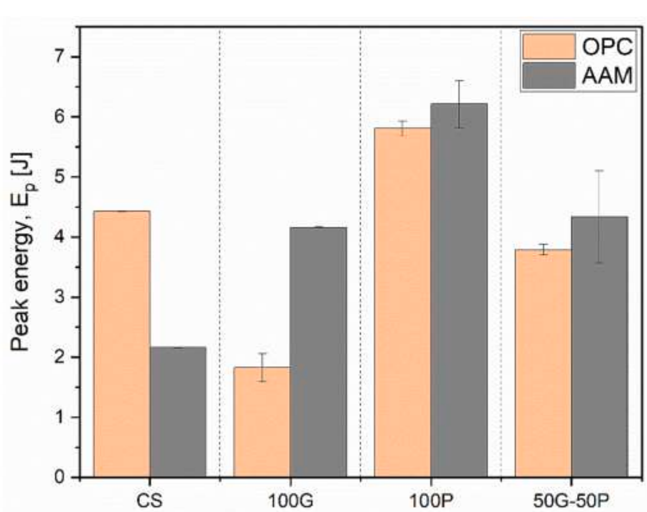

Fig. 8. Impact peak energy (E_p) absorption of the composites.

To support the dynamic mechanical test results, Fig. 9 displays the failure modes of the sample after impact load. The extent of surface damage clearly reflected the microstructural quality and, therefore, the energy absorption capacity of the investigated composites. At the same sand replacement level with the two recycled aggregates (waste glass and PVC), a lower entity of damage was detected in the AAM samples compared to OPC, justifying the better compatibility of the waste aggregates with the binder and, therefore, their functionality in terms of impact behaviour.

3.2.2. Thermal conductivity

In a steady-state heat transfer scenario, thermal conductivity depends on the material's properties, surface area, thickness, and temperature gradient [49]. As illustrated in Fig. 10, all AAM composites attained lower thermal conductivity than their OPC counterparts, which is attributed to the higher thermal stability of calcium-aluminate-silicate-hydrate (C-A—S—H) products in AAMs than the calcium silicate hydrate (C—S—H) in OPC. The AAMs matrix was more resistant to high temperatures than the OPC matrix [50]. This has been verified in the current study, where the thermal conductivity of the AAM-CS (i.e., 2.5 W/mK) was lower than the OPC-CS (i.e., 2.75 W/mK). In both AAM and OPC-based composites, the induction of recycled PVC and glass aggregates resulted in a substantial reduction in the composites' thermal conductivity. The thermal conductivity reduction is due to the lower thermal conductivity of individual recycled PVC (i.e., 0.17 W/mK) [29] and waste glass 0.93 (W/mK) [30] aggregates compared to that of natural sand aggregates (i.e., 3–3.5 W/mK) [31]. As a result of the lower thermal conductivity of PVC aggregates than waste glass aggregates, composites incorporated with PVC aggregates exhibited higher thermal conductivity reduction than those with glass aggregates. The maximum thermal conductivity reduction was recorded for AAM-100P and OPC-100P composites with a reduction of about 84 % and 78 %, respectively, compared to their corresponding control samples. As previously mentioned, (see Section 3.1), the addition of recycled aggregates results in the generation of air-filled gaps between the aggregates and matrix. The entrapped air with thermal conductivity of roughly 0.026 W/mK at 20 °C [36] acts as a thermal insulator and prevents heat transfer through the cementitious composites' structure, consequently reducing the overall composites' thermal conductivity. These findings corroborated with the study of Latroch et el. [51], where recycled PVC was used at 75

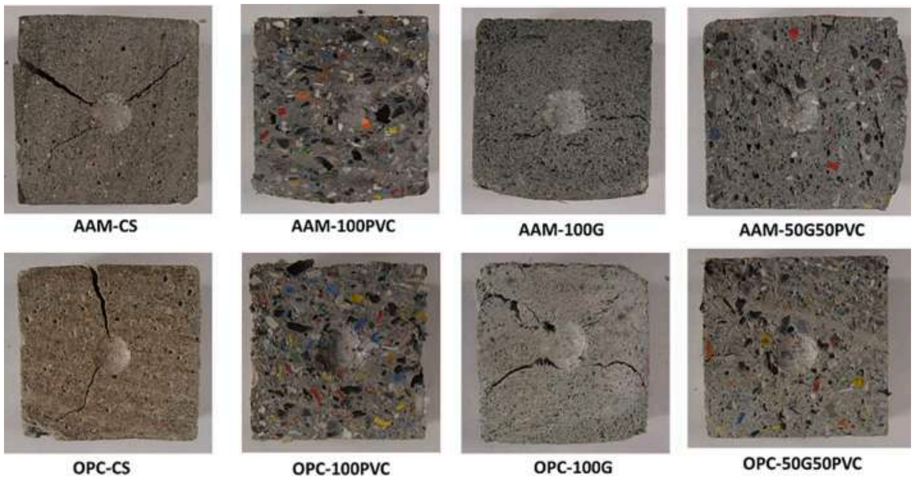

Fig. 9. Failure patterns of the composites after the impact test.

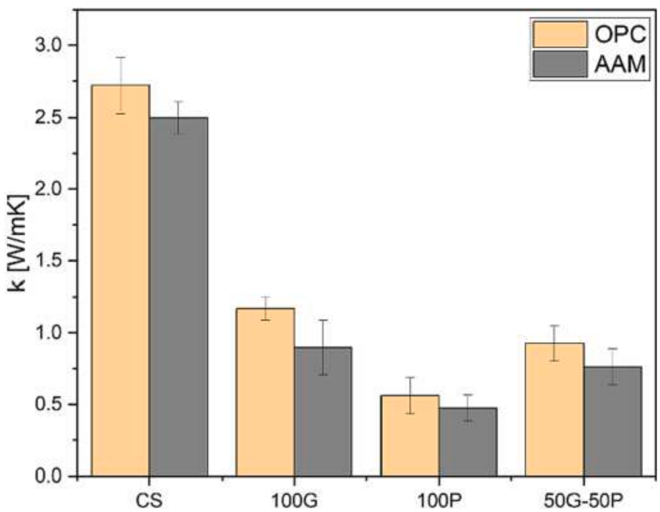

Fig. 10. Thermal conductivity of the composites.

% replacement with sand, and thermal conductivity was reduced by 60 % compared to OPC-based composites with 100 % natural sand. Khalil et al. [52] concluded that due to the unspecific recycled plastics aggregate's lower thermal conductivity than natural aggregate, the overall thermal conductivity of the incorporated composites dropped.

3.2.3. Water absorption and harsh environment resistance

Due to the higher performance of AAM composites than OPC-based mixes in terms of mechanical and physical characteristics, AAM samples were only selected to investigate their water absorption and resistance to harsh environments. The effect of aggregate substitution on the water uptake behaviour of AAM composites is shown in Fig. 11a. The results indicated that the water absorption of the tested samples drastically increased after four days (i.e., 5760 min) of immersion in water, rising from 3.5 % for the control sample to 5 %, 4.6 %, and 4.5 % for the 100P, 100G, and 50G-50P, respectively. A previous study [53] has reported a significant increase in the water absorption capacity of cementitious composites due to an increase in the natural sand replacement ratio. The effect is linked to the inadequate mixing of the cementitious matrix with recycled plastic aggregates, leading to the generation of trapped air in the fresh mixture and consequently increasing the resulting composite's porosity. Albano et al. [30] reported that the other explanation could be linked to the content, incompatibility, size, and shape of recycled plastic aggregates, which

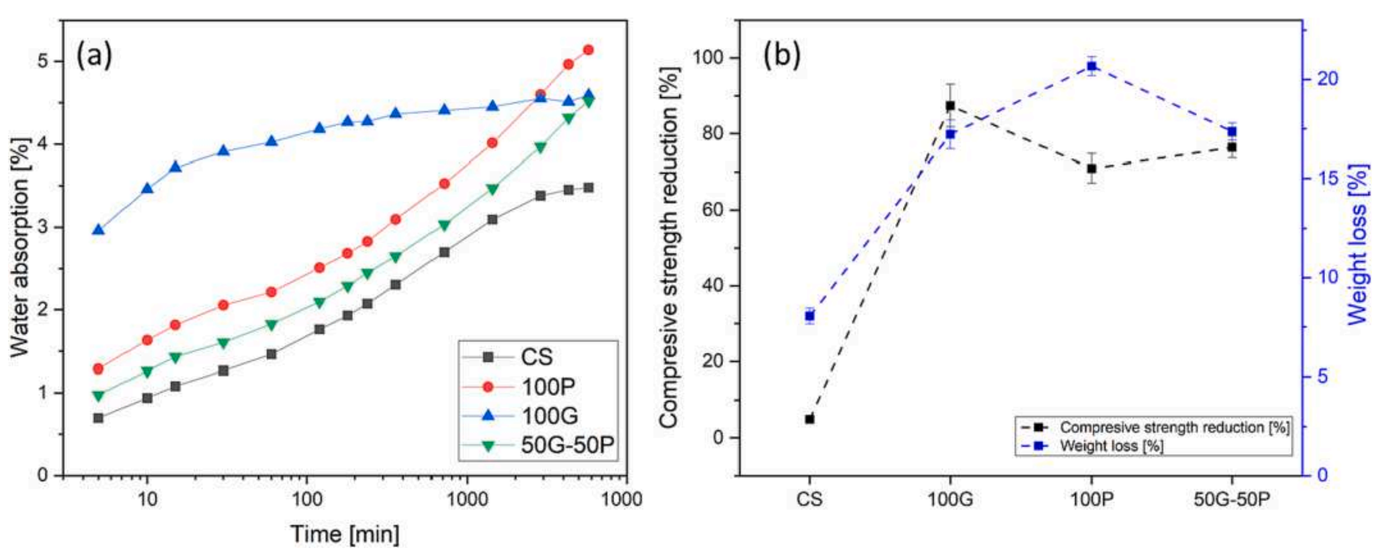

Fig. 11. (a) Water absorption, and (b) harsh environment resistance of the alkali-activated composites.

directly influence the porous structure of composites by altering the cementitious paste quality. The results in Fig. 11a also indicated that water absorption progressively increased with a high slope over time for the CS, 100P, and 50G-50P samples. However, in the case of the 100G mixture, a high-water absorption level was observed immediately after immersion, followed by a lower increment over time. Glass is an impermeable material [54], however, the results suggest that the presence of glass aggregates as a substitute for natural aggregates in AAMs increases the composites' water absorption. As seen in the microstructural analysis of the samples containing glass aggregates (see Fig. 7b), the poor interfacial bonding between glass aggregate and AAM matrix facilitates the water transport through the samples and consequently increases water uptake.

The resistance of AAM composites to harsh environmental conditions was also assessed by recording the loss in their weight and compressive strength values at the end of 10 cycles of harsh conditions. In each cycle, samples were frozen for 24 h at −14 °C and then thawed in water at +70 °C for the next 24 h. As shown in Fig. 11b, all AAM composites containing recycled aggregates (i.e., 100P, 100G, and 50G-50P) experienced a substantial deterioration when exposed to the harsh environment, i.e., 71 % – 87 % compressive strength reduction and 17 % – 20 % weight loss. According to a study conducted by Ferrándiz-Mas and García-Alcocel [55], plastic aggregates can partially relieve the crystallisation pressure of ice-freezing, which diminishes composites' deterioration at certain replacement levels. Nevertheless, their findings suggested that when plastic aggregates' replacement level exceeded 50 %, there was a substantial weight loss and compressive strength reduction. In this study, the ice-induced deterioration of samples containing PVC aggregates could be due to: (i) poor workability in the fresh state [56] and (ii) weak matrix-aggregate interfacial bonding (observed in microstructural analysis in Fig. 7b and d) of mixtures containing recycled plastic aggregates, which increases the porosity of the resultant AAM composites, therefore making it easier for freezing-induced microcracks to develop. Furthermore, Kan and Demirboa [57] concluded that the ice-freezing deterioration degree of cementitious composites containing recycled plastic is also related to the size of the employed plastic aggregates, where coarse plastic aggregates make cementitious composites more vulnerable to ice-freezing deterioration than fine ones. As seen in the microstructural analysis of Fig. 7c, the same statement, i.e., weak aggregate-matrix interlocking and increased porosity of AAMs containing recycled aggregate, can be employed to elucidate the extreme ice-induced damages in the 100G composite.

3.3. Carbon footprint estimation

The environmental impact assessment was calculated through the SimaPro software using the IPCC 2021 method, which provides the carbon footprint of the selected processes in the Global Warming category, evaluated in kg of CO_2 equivalent. Table 2 shows the environmental impact values per kilogram of component (EC) and the corresponding weight (m) in kilograms for one ton of mixture. The negative EC-value detected for glass recycling process, as well as for PVC, can be explained by referring to the LCA networks (Figs. 3 and 4) built by SimaPro. Comparing the recovery processes of the waste material for use in the AAM with the carbon footprint associated to their grinding process to obtain the granular fraction, two rates of environmental impact are observed. Recovery involves "negative" EC values (−0.883 kg CO2-eq for glass and −1.89 kg CO2-eq for PVC), as using PVC and glass aggregates would avoid the eco-impact associated with the production process of the virgin raw materials. On the other hand, the material's processing implies direct carbon emissions, therefore "positive" EC-values related to the consumption of electrical energy (0.00761 kg CO2-eq for glass and 0.23 for PVC) and other resources (0.00158 kg CO2-eq for natural gas in waste glass processing) required for grinding. By summing the two contributions, the EC-values of PVC and glass aggregates in Table 2 can be obtained.

The assessment of the carbon footprint of cementitious mixtures (i) is cumulative to the impacts of the individual components (j) and was obtained using Eq. (2):

$$GWP_i = \sum_j m_{i,j} \times EC_j$$

Where GWP_i is the global warming potential of the i-esimal mixture. Fig. 12 shows the results of the GWP calculation for the four different compositions of the OPC and AAM mixtures.

Comparing the impacts of the two control samples, it is evident that the carbon footprint of the OPC (260.79 kg CO_2-eq) is greater than that of the AMM mixture (144.77 kg CO_2-eq), confirming the advantage in terms of carbon emissions resulting from the use of AAM binders [33,58]. By replacing PVC aggregates with sand, there is a reduction in the impact due to the negative contribution of the PVC recovery process and, overall, an environmental benefit is obtained, in terms of carbon footprint, both for AAM (−352.25 kg CO_2-eq) and OPC (−199.79 kg CO_2-eq). This result agrees with the research conducted by Imteaz et al. [59], in which the authors demonstrated that if recycled PVC is used instead of virgin material in railway concrete applications, a carbon footprint savings more than 50 kg CO2-eq can be achieved. Similarly, by

Table 2
Environmental impact values (ECs) of the AAM and OPC-based mix designs.

Components	mCS (kg)	m100P (kg)	m100G (kg)	m50G-50P (kg)	EC (kg CO_2-eq/kg)	EC's Reference
AAM						
FA	223.3	283.1	248.5	239.9	0.0213	[31]
GGBS	93.0	117.9	103.5	100.0	0.0729	[31]
SF	55.8	70.8	62.1	60.0	0.0140	[33]
F SAND	278.7	–	–	–	0.00471	[31]
C SAND	185.8	–	–	–	0.00471	[31]
Sodium silicate	99.2	125.8	110.5	106.6	0.705	[31]
Sodium hydroxide	49.6	62.9	55.2	53.3	0.777	[31]
PVA fibre	10.9	13.8	12.1	11.7	2.00	[34]
PVC aggregate	–	321.1	–	96.7	−1.66	[4]
Glass aggregate	–	–	404.0	327.9	−0.874	This study
Attapulgite	3.6	4.6	4.0	3.9	–	–
OPC						
OPC	283.3	423.1	231.3	297.6	0.851	[31]
Water	141.6	211.6	115.7	148.8	0.000324	[31]
Sand	566.6	–	–	–	0.00471	[31]
PVA fibre	8.5	12.7	6.9	8.9	2.00	[34]
Glass aggregate	–	–	646.1	420.6	−0.874	This study
PVC aggregate	–	352.6	–	124.0	−1.66	[4]

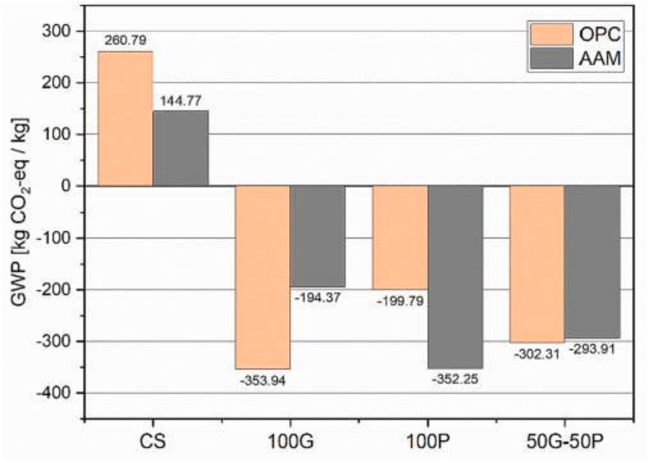

Fig. 12. Carbon footprint estimation (GWP) of the composites.

fully replacing sand with glass aggregates, carbon footprint values of −194.37 kg CO_2-eq for the AAM and −353.94 kg CO_2-eq for the OPC are obtained. The beneficial environmental effect of using waste glass in concrete is corroborated by the results reported in [60]. If the volume of sand is replaced half by PVC and half by the glass, environmental impact values are obtained (−293.91 kg CO_2-eq for the AAM and −302.31 kg CO_2-eq for the OPC) between the respective values of PVC and glass. In both cases, a net environmental benefit is also obtained. In other words, the GWP depends on the type of binder, type of waste material used as aggregate replacement, and its weight content in the mix designs [61]. For 100G sample, OPC matrix admitted the higher waste glass content thus implying a greater environmental gain than the AAM counterpart (GWP of cementitious composite about twice lower than AAM one). However, as previously verified, high dosage of glass aggregates in Portland-based concrete clashed with lower static-dynamic mechanical performances and heat resistivity compared to the alkali-activated binder. A reverse trend in terms of carbon footprint value was obtained by totally incorporating PVC aggregates in the AAM system (100P-AAM). In this case, however, the polymer waste addition brought improvement on the material's behaviour, specifically on flexural strength, mechanical dynamic response, and thermal insulation. Although the AAM mix design incorporated less weight content of PVC than the OPC mix design, the environmental benefit of the matrix would seem to further lower the overall carbon footprint of the composite. BY designing the "hybrid" mixtures, with incorporating equal contents of glass and PVC aggregates, we balanced the environmental impact of the two composite systems. Furthermore, as verified in Section 3.1, the same effect was also found in the mechanical performances. It is therefore interesting to deduce that, by balancing the content of the two waste fractions, almost binder-independent eco-footprints and technological performances can be achieved.

4. Conclusions

Recycled PVC and glass aggregates could be used to develop lightweight, low-carbon, alkali-activated cementitious composites. The microstructure, mechanical performance, and physical characteristics of cementitious composites with and without recycled aggregates were comprehensively examined. For all AAM and OPC-based composites, the density of the composites was reduced using recycled PVC and glass aggregates.

- AMMs are superior to OPC-based composites in terms of mechanical performance at all replacement ratios. The highest compressive strength (i.e., 30 MPa) was registered for 100G and the highest flexural strength (9 MPA) for 100P. Apart from 100 G in OPC-based composites, energy absorption in AMMs was improved by replacing natural sand with recycled PVC and glass. The AAM-100P had a peak energy absorption of roughly 6 J, which is three times greater than the AAM-CS.
- Recycled PVC and glass aggregates were used to enhance the thermal resistance of composites. AAM-100P exhibited the lowest thermal conductivity with a value of 0.5 (w/mK), which is five times less than AAM-CS. However, samples with PVC aggregates demonstrated superior heat resistance compared to samples with glass.
- The 100P and 50G-50P composites exhibited the lowest compressive strength and weight reductions, at about 70 % and 15 %, respectively. Due to the weak interlock between the recycled aggregates and matrix, the addition of PVC and glass aggregates increased the water absorption rate of AAMs. The structural and mechanical characteristics of composites containing AAM degraded when exposed to a hostile environment.
- In terms of dynamic mechanical properties, replacing the sand with the two recycled fractions in AAM composites increased energy absorption. Superior performances were found in the samples totally replaced with PVC aggregates (AAM-100P).
- The 100 % replacement of sand with PVC or glass or the hybrid (i.e., 50G-50P) replacement reduces the overall carbon footprint of mixtures and results in a net benefit in the environmental category of global warming. For the AAM mixtures, a more significant reduction of the environmental impact is obtained by replacing the sand with PVC, while for the OPC, the most significant environmental benefit is obtained by replacing the waste glass with the sand.

The most important results of replacing natural sand with glass and PVC are a conduit for recycling waste products; an improvement in composites' thermal resistance; an aid in reducing the shortage of natural aggregates; a reduction in the building industry's carbon footprint; and few applications such as Cladding tiles, highway sound barriers, pavement stones, parking for energy and sound absorption.

Future research directions include using different types of recycled PVC, such as rigid PVC, and testing different combinations of PVC and glass aggregates to get the best mechanical and physical performance of the composites. Moreover, working with different sizes of PVC and glass aggregates to fill the matrix pores will improve the bond between the aggregates and paste, and using PVC and glass aggregates in one-part geopolymers will result in a lower carbon footprint.

The findings of this study show that, despite recycled PVC and glass aggregates' detrimental effects on AAMs' mechanical performance, which restrict their use in load-bearing structures, these lightweight composites possess good energy absorption and thermal insulation properties. These characteristics could potentially be an incentive to help preserve natural resources, reduce plastic pollution, and recycle aluminosilicate solid waste to lower CO_2 emissions. Yet, concerning the material's particular applications and operating circumstances, additional research on the durability of AAMs comprised of recycled PVC and glass aggregates is required. Further research in finding efficient methods to modify the surface of recycled aggregates to enhance the interface between the aggregate and matrix, reducing the loss of mechanical properties, is also of interest.

CRediT authorship contribution statement

Eslam El-Seidy: Data curation, Methodology, Validation, Investigation, Writing – original draft, Visualization. **Mehdi Chougan:** Data curation, Writing – review & editing, Data curation. **Matteo Sambucci:** Data curation, Writing – review & editing, Data curation. **Mazen J. Al-Kheetan:** Writing – review & editing, Data curation. **Ilario Biblioteca:** Writing – review & editing, Data curation. **Marco Valente:** Writing – review & editing, Validation. **Seyed Hamidreza Ghaffar:** Conceptualization, Methodology, Supervision, Writing – review & editing, Resources, Writing – original draft, Visualization, Supervision, Funding acquisition.

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.

Data availability

Data will be made available on request.

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