One-part alkali-activated materials: A review

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ABSTRACT

Alkali-activated materials (AAM) are recognized as potential alternatives to ordinary Portland cement (OPC) in order to limit CO_2 emissions as well as beneficiate several wastes into useful products. However, the alkali activation process involves concentrated aqueous alkali solutions, which are corrosive, viscous, and, as such, difficult to handle and not user friendly. Consequently, the development of so-called one-part or "just add water" AAM may have greater potential than the conventional two-part AAM, especially in cast-in-situ applications. One-part AAM involves a dry mix that consists of a solid aluminosilicate precursor, a solid alkali source, and possible admixtures to which water is added, similar to the preparation of OPC. The dry mix can be prepared at elevated temperatures to facilitate the reactivity of certain raw materials. This review discusses current studies of one-part AAMs in terms of raw materials, activators, additives, mechanical and physical properties, curing mechanisms, hydration products, and environmental impacts.

1. Introduction

Ordinary Portland cement (OPC) contributes significantly to the global CO_2 emissions: in 2016, the estimated value was 1.45 ± 0.20 Gt CO_2, that is, approximately 8% of the total anthropogenic CO_2 release [1]. Although the cement industry has been able to improve energy efficiency significantly, increasing cement demand (estimated as 3.36–3.48 Gt in 2015 and 3.68–4.38 Gt per year by 2050) has outpaced much of the positive impact [2]. Furthermore, approximately 50–60% of OPC-production-related CO_2 emissions are released from the calcination (decarbonation) of limestone at 1400–1450 °C (Reaction 1), which cannot be influenced by improving energy efficiency [3,4].

$$CaCO_3 \text{ (s)} \rightarrow CaO \text{ (s)} + CO_2 \text{ (g)} \qquad (1)$$

Consequently, the development of alternative low-carbon binders is recognized as one option to reduce CO_2 emissions [5,6]. Geopolymers, which are sometimes considered a sub-group of alkali-activated materials (as originally suggested by Irene Beleña) are promising materials in this regard [7]. While they are unlikely to completely replace OPC, they may serve as an alternative and supplementary binder depending on the local availability of raw materials [8]. Alkali-activated materials could be designed to have superior properties compared to binders prepared from OPC, namely better resistance to acids and sulfate [9–11], better heat resistance [12–14], lower drying shrinkage and creep [15], and higher strength [16]. However, efflorescence formation can be a potential problem if not properly controlled by, for example,

mix design [17]. In terms of unwanted alkali-aggregate reactions, alkali-activated materials have frequently showed better performance than OPC, as documented, for instance, by García-Lodeiro et al. [18] but attention should be paid for the proper selection of type and dose of activator, type of binder, and type of aggregates in this regard [19]. Alkali activation technology also allows a waste beneficiation route for utilizing several industrial by-products [20]. In addition, many of the current standards for cementitious materials do not recognize the use of alkali-activated materials as they have been written from the viewpoint of OPC use (i.e., they include prescriptive compositional limits instead of performance-based specifications) [21]. However, in some countries, such in Ukraine [22], the use of alkali-activated cements is already included in the legislation. Also, supply chains for raw materials, suitable admixtures for alkali-activated materials, and testing protocols are still inadequate [23].

Binders prepared by reacting an alkali source and solid aluminosilicate were patented as early as 1908 [24]. Later, important pioneering work was done by Glukhovsky [25], Krivenko [26], Davidovits [27], and Palomo et al. [28] among many others. A complete historical overview of alkali-activated materials is available in several reviews [29–32]. However, there is still some ongoing debate about the terminology related to alkali-activated materials and geopolymers: the former is generally characterized by a lower coordination of Si, that is, Q^2 and $Q^2(1Al)$ and higher calcium content, whereas the latter contains more $Q^4(2Al)$ and $Q^4(3Al)$ centers, has low calcium content, and is essentially three-dimensional [7,33–35]. However, the terms
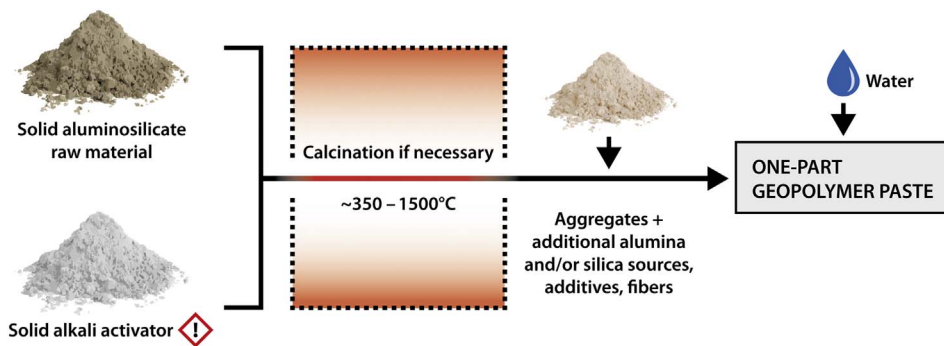
Fig. 1. The general procedure of one-part geopolymer preparation.

geopolymer and alkali-activated material are used in the literature somewhat interchangeably and occasionally even as synonyms. In this review, materials are referred to as using the same terminology as used in the cited references.

Conventional (two-part) geopolymers are formed by a reaction between a concentrated aqueous solution of alkali hydroxide, silicate, carbonate, or sulfate, for instance, and solid aluminosilicate precursor, that is, two parts in addition to water [36–40]. However, the impracticalities related to handling large amounts of viscous, corrosive, and hazardous alkali activator solutions has put pressure on the development of one-part or "just add water" geopolymers that could be used similarly to OPC [41]. In one-part mixtures, only a dry mixture is needed in addition to water. The dry mixture is prepared by mixing a solid alkali-activator with a solid aluminosilicate precursor with or without a calcination step (Fig. 1).

In 1940, Purdon [42] proposed dry mixing of slag and solid sodium hydroxide and subsequently adding water in order to prepare a mortar mixture. In the 1980s, Heitzmann et al. [43] patented a dry mixture of metakaolin, blast furnace slag, amorphous silica, potassium hydroxide and silicate, and one of the following components: fly ash, calcined shale, or calcined clay that could be blended with OPC before the addition of water. Schwarz and Andre [44] patented a geopolymeric dry mixture in which amorphous silica was prepared by dealuminating fly ash or metakaolin with hydrochloric or sulfuric acid. Davidovits [45] also patented one-part geopolymer consisting of alumino-silicate oxide with Al in IV-V coordination (i.e., metakaolin), sodium or potassium disilicate, and slag. Later, Davidovits [46] pointed out that solid sodium or potassium hydroxide and silicate should be partially replaced with "synthetic lavas" (reactive sodium-potassium aluminosilicate glass) due to large-scale availability and production issues. Synthetic lava could be prepared by mixing volcanic tuff and calcium carbonate, melting at 1200–1350 °C, quenching in water, drying, and grinding [46]. Duxson and Provis [41] outlined some general needs and approaches for one-part geopolymer mixes. They proposed that solid precursor would be prepared either by adding, for instance, calcium-containing feldspar to coal before combustion; melting the previous materials separately and grinding; or preparation of a two-part geopolymer as a precursor to a one-part geopolymer. Currently, one-part geopolymers are considered especially promising for in situ applications where handling alkali solutions can be difficult whereas two-part mixtures appear suitable for precast work [8,23,47]. However, the early examples of one-part geopolymers described above have attained little commercial use. In contrast, two-part geopolymers have already been used in several full-scale implementations, such as the Brisbane West Wellcamp Airport in Australia [48].

The purpose of this review is to present the current state-of-art in the development of one-part geopolymer mixtures. A substantial number of review articles exist discussing various aspects and applications of two-part geopolymers [49–61], but no similar reviews about one-part mixtures currently exist. This paper discusses the employed aluminosilicate precursors, solid activators, admixtures, mix designs,

resulting binding phases, hardening mechanisms, and effects of various parameters on the mechanical properties of one-part geopolymers. Moreover, a brief overview of one-part geopolymers containing OPC as one component (i.e., hybrid cements or blended alkaline cement) is presented. Finally, the environmental impact and cost analysis of one-part mixes is provided.

2. Raw materials and preparation of one-part geopolymers

2.1. Aluminosilicate precursors

The most common solid aluminosilicate precursor in one-part geopolymer mixes is fly ash from coal combustion either alone or in combination with blast furnace slag (Table 1). The majority [62–69] of the fly ashes used as precursors fall within class F (low calcium content) as defined by ASTM standard C618 [70]. Class C (high calcium content) fly ash is less frequently used in geopolymer binders because of too rapid setting [71,72] and less abundant availability [73]. Ye et al. [74] used one-part geopolymer approach on the successful solidification/stabilization of municipal solid waste incineration fly ash (containing metals such as Pb, Cu, Zn, and Cr) by mixing it with red mud, NaOH, and then adding water. While fly ashes have usually been employed without pretreatment in one-part mixes, Matalkah et al. [75] used a mechanochemical activation method in which fly ash was ball-milled with dry-blended activators (CaO, MgO, and NaOH). They observed that Na, Ca, and Mg were incorporated in the fly ash structure due to disruption of the aluminosilicate bonds, which resulted in improved properties (higher strength, greater moisture resistance, and finer microstructure) compared to raw materials that were separately milled and then blended [75]. Similar mechanochemical pretreatments have been used with two-part geopolymers as well with a significant increase in compressive strength [76] and rate of geopolymerization [77].

Ground-granulated blast-furnace slag is commonly used as a calcium-rich aluminosilicate precursor in alkali-activated materials. Blast furnace slag, which is a by-product of pig iron manufacturing at blast furnaces, consists of silicate and aluminate impurities present in iron ore and coke. Blast furnace slag is frequently mixed with class F fly ash in one-part geopolymer mixtures, but it can also be used alone (Table 1). Using blast furnace slag improves the reactivity of low-calcium-content fly ash [41]. Usually calcium-rich aluminosilicate or calcium hydroxide would induce rapid setting and high early strength [64,78], but replacement of a portion of the slag with calcium hydroxide resulted in a decrease in both compressive strength and workability in a one-part geopolymer [65]. Decreasing the particle size of blast furnace slag resulted in an increase of compressive strength [63]. Wang et al. [66] found that a ratio of blast furnace slag to solid water glass of 5:1 (by weight) was optimum in terms of compressive strength, whereas ratios of 4:1 and 6:1 led to decreased strength. Nematollahi et al. [67] used "typical" and "gypsum-free" slags in the design of one-part strain-hardening geopolymer composite. The use of "typical slag" resulted in a lower relative slump value, significantly higher thixotropic

Table 1
One-part alkali-activated material studies reported in the literature: raw materials; admixtures; optimum calcination, compositions, and curing conditions; and obtained unconfined compressive strengths.

(Alumino) silicate precursor	Solid activators	Admixtures and fibers	Aggregates	Calcination	Optimum molar ratios						L/S^a	Curing		7 d UCS [MPa]	14 d UCS [MPa]	28 d UCS [MPa]	Ref.
					SiO_2/Al_2O_3	Na_2O/SiO_2	Na_2O/Al_2O_3	$(Ca+Mg)O/SiO_2$	H_2O/Na_2O	H_2O/Al_2O_3		T [°C]	RH [%]				
Fly ash	$(Na_2SiO_2)_nO$, NaOH	–	Std. quartz sand	–	1.8^c	0.8^b	1.5	n.r.	12	18^b	0.25	40	n.r.	≈42	≈57	n.r.	[69]
Fly ash	CaO, NaOH, MgO	–	Natural sand and granite	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.40	23	100	≈17	≈25	≈35	[75]
Fly ash (LOI > 6%)	Red mud or NaOH	–	–	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.34–0.56	23 + 60	n.r.	≈1.6	≈1.6	≈1.6	[98]
Fly ash	$Ca(OH)_2$, Na_2SiO_3	PVA fibers	–	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.20	23	n.r.	n.r.	n.r.	n.r.	[139]
Fly ash	NaOH, $NaAlO_2$, Al_2O_3	–	–	–	0.75^b	n.r.	1^b	n.r.	12	n.r.	0.338	40	n.r.	≈45	≈50	≈55	[161]
Fly ash	Na_2SiO_3	PVA fibers	–	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.35	23	n.a.^{c}	n.r.	n.r.	48.7	[136]
Fly ash	Na_2SiO_3	–	–	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.40	20	100	2.35	6.04	11.29	[128]
BFS, fly ash	Na_2SiO_3	PVA fibers	–	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.35	23	n.a.^{c}	n.r.	n.r.	52.5	[67]
BFS, fly ash	Na_2SiO_3	PE or PVA fibers	–	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.35	23	n.a.^{c}	n.r.	n.r.	48.7	[68]
BFS, fly ash	$(Na_2SiO_2)_nO$	Hydrophosphate (retarder)	–	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.28	n.r.	n.r.	67.38	n.r.	80.13	[66]
BFS or fly ash	$(Na_2SiO_2)_nO$, NaOH	–	Sand	–	n.r.	1.2–1.8^b	n.r.	n.r.	n.r.	n.r.	0.5	23	70	38.5	n.r.	49.6	[63]
BFS, fly ash	Na_2SiO_3, NaOH, $Ca(OH)_2$	PCE	–	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.3	23	n.a.^{c}	33.9	n.r.	36.9	[65]
BFS or fly ash	$(Na_2SiO_2)_nO$	–	Sand	–	5.08^b	0.26^b	1.31^b	1.29^b	19.01^b	24.94^b	0.50	23	70	47.08	n.r.	51.28	[62]
BFS or fly ash	$(Na_2SiO_2)_nO$	–	Sand	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.3	23	70	64.5	n.r.	71.6	[162]
BFS	NaOH, Na_2CO_3	–	–	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.27	37	100	n.r.	n.r.	n.r.	[163]
BFS	CaO or $Ca(OH)_2$	–	–	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.40	25	99	31	34	42	[113]
BFS	$(Na_2SiO_2)_nO$	Polycarboxylic-based water reducing admixture	Expanded clay granule, sand	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.4	n.r.	n.r.	49.3	n.r.	53.8	[127]
BFS, silica fume	Na_2CO_3, slaked lime	Sodium lignosulfonate	Dolomite sand and stone	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.35	25	> 90	n.r.	n.r.	≈50	[117]
Fayalite slag	Maize cob ash	–	Std. quartz sand	–	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.28	20	100	n.r.	n.r.	16.4	[80]
Rice hull ash	$NaAlO_2$	–	Std. quartz sand	–	1.25^b	0.98^b	1.27^b	n.r.	14	17.78^b	0.308	40	100	≈8	≈18	n.r.	[102]
Rice husk ash	$NaAlO_2$	–	–	–	3.48	0.29^b	1.00	n.r.	11.85^b	11.81	0.5	80	80	30.1	n.r.	n.r.	[103]
Bentonite	Dolomite, Na_2CO_3	–	–	1100 °C/3 h	4.6	0.4	1.84^b	2.1	n.r.	n.r.	0.35	80 + 20	100	n.r.	n.r.	38.3	[152]
Bentonite	NaOH or Na_2CO_3	–	–	850 °C/3 h	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.30	80 + 20	100	≈30	n.r.	≈25	[116]
Albite	NaOH or Na_2CO_3	–	–	1000 °C/0.5 h	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.3	25	100	32.3	38.5	44.2	[85]
Kaolin	NaOH or Na_2CO_3	–	–	950 °C/3 h	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.3	80 + 25	> 90	n.r.	n.r.	n.r.	[115]
Kaolin	NaOH, KOH	–	–	550 °C/4 h	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	≈23	n.r.	≈1	n.r.	n.r.	[153]

(continued on next page)

Table 1 (continued)

(Alumino)silicate precursor	Solid activators	Admixtures and fibers	Aggregates	Calcination	Optimum molar ratios						L/S^a	Curing		7 d UCS [MPa]	14 d UCS [MPa]	28 d UCS [MPa]	Ref.
					SiO_2/Al_2O_3	Na_2O/SiO_2	Na_2O/Al_2O_3	$(Ca+Mg)O/SiO_2$	H_2O/Na_2O	H_2O/Al_2O_3		T [°C]	RH [%]				
Kaolin	Maize cob ash	–	Std. quartz sand	700 °C/1 h	3.07	0.76^b	1.28^d	n.r.	n.r.	n.r.	0.3^b	20 + 80	60^e	40	n.r.	n.r.	[105]
Metakaolin	NaOH, $(Na_2SiO_2)_nO$	H_2O_2, surfactant	Std. quartz sand	–	1.75^b	0.88^b	2	n.r.	14	28^b	0.75^b	60 + 20	100	≈ 45	≈ 65	n.r.	[158]
Silica residue, silica fume	$NaAlO_2$	–	–	–	6.02	0.16^b	0.98	n.r.	15.82^b	15.51	0.34	80 + 23	80 + 50	n.r.	n.r.	n.r.	[106]
Silica residue or silica fume	$NaAlO_2$	–	–	–	5.35	0.18^b	0.98	n.r.	14.64^b	14.36	0.50	80	80	n.r.	n.r.	n.r.	[107]
Silica residue	$NaAlO_2$	–	–	–	3.5	2.92^b	1.2^b	n.r.	11.3	n.r.	0.60	70 + 23	100	≈ 7	n.r.	n.r.	[108]
Geothermal silica	$NaAlO_2$	–	–	–	$0.75–1.2^b$	$3.00–4.99^b$	1.00^b	n.r.	7–12	$7–12^b$	0.214–0.386	40	100	n.r.	n.r.	n.r.	[110]
Geothermal silica	$NaAlO_2$	Al_2O_3, ZnO, or ZrO_2	Sand	–	0.75^b	n.r.	n.r.	n.r.	7	n.r.	n.r.	40	100	≈ 22	≈ 26	n.r.	[135]
Red mud	NaOH	–	–	800 °C/1 h	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.50	20	100	≈ 2.5	≈ 2.2	≈ 1.8	[74]
Red mud, silica fume	NaOH	Lignosulfonate	–	800 °C/1 h	3.45	0.42	n.r.	n.r.	12.96	n.r.	0.45	20	100	≈ 13	n.r.	31.5	[97]
Red mud	NaOH	–	–	800 °C/1 h	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.60	20	95	10	n.r.	≈ 2	[96]
Potash feldspar, albite, clay, pyrophyllite, kaolin, natural zeolite, fly ash, BFS	KOH, NaOH, Na_2CO_3, Ca(OH)$_2$, $CaSO_4$, OPC	Several oxidants, setting control agents, and retardants mentioned	Sand and other fillers	350–1500 °C	1	1^b	1	$0.02–0.2^b$	n.r.	n.r.	0.35	20	100	4.5–32.5	7.1–39.5	10.2–45.3	[64]
OPC, and fly ash and bottom ash	$CaSO_4$ and Na_2SO_4	–	Std. sand	–	6.33	0.33	2.12	3.56	n.r.	n.r.	0.5	21	99	20	n.r.	32.5	[145]
OPC, BFS, metakaolin	Na_2CO_3	–	–	750 °C/2 h	3.61	0.01	0.03	0.85	n.r.	n.r.	0.5	22	99	15 (2 d)	n.r.	n.r.	[147]
OPC, BFS, metakaolin	Na_2CO_3	–	–	750 °C/2 h	6.51	0.01	0.11	0.80	n.r.	n.r.	0.3	22	99	32 (2 d)	n.r.	n.r.	[147]
OPC, fly ash, kaolin	Ca(OH)$_2$, NaOH, KOH	Lignosulfonate	Sand	6500 °C/140 min	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.29–0.4	n.r.	n.r.	8–22	20–27	16–42	[148]
OPC, fly ash, kaolin	Ca(OH)$_2$, NaOH	Lignosulfonate	Sand	6500 °C/140 min	n.r.	n.r.	n.r.	n.r.	n.r.	n.r.	0.35	n.r.	n.r.	n.r.	12–25	15–27	[150]

n.a. = not applicable; n.r. = not reported; $(Na_2SiO_2)_nO$ = general formula for sodium silicate; PVA = polyvinyl alcohol; PE = polyethylene; PCE = polycarboxylate ether; BFS = blast furnace slag; UCS = unconfined compressive strength; OPC = ordinary Portland cement.

a Liquid to solid, i.e., water to binder, ratio.
b Calculated from the provided data.
c Cured under water.
d $(Na_2O + K_2O)/Al_2O_3$.
e Relative humidity at 80 °C not reported.

property, higher elastic modulus, less brittle structure (indicated by higher fracture toughness and crack tip toughness), and higher uniaxial tensile performance [67]. This indicates that the presence of gypsum in slag is unfavorable.

Fayalite slag, on the other hand, forms in the secondary copper or nickel smelting process as SiO_2 is introduced into the smelter [79]. The amount of fayalite slag generated is approximately 2.2 tons per one ton of produced copper [79]. It consists mainly of iron and zinc silicates, $(Fe,Zn)_2SiO_4$, but also contains PbO, Al_2O_3, and CaO [79]. Fayalite slag was recently utilized in a one-part geopolymer in which it was dry mixed with maize cob ash (weight-ratio ash/slag of 0.4) to which water was added [80]. The hardened material reached rather modest compressive strength of 16 MPa at the age of 28 d (Table 1).

Clay minerals (hydrous aluminosilicates) typically require calcination in order to be reactive in alkali activation processes. Their plate-like particle shape increases water demand and subsequently results in concrete with high porosity [41]. Kaolin $(Al_2Si_2O_5(OH)_4)$ in its dehydroxylated form, metakaolin $(Al_2Si_2O_7)$, is one of the most studied aluminosilicate sources for geopolymer preparation. Bentonite, another potential geopolymer precursor, usually consists mainly of the clay mineral montmorillonite, $(Na,Ca)_{0.33}(Al,Mg)_2(Si_4O_{10})(OH)_2 \cdot nH_2O$. In alkali activation, however, bentonite shows a low reactivity as such [81], but it can be improved by thermal treatment at 550–850 °C [82–84]. Van Deventer et al. [64] listed additional clays and other natural minerals that could be used as an aluminosilicate source in one-part geopolymers such as albite (sodium feldspar, $Na(AlSi_3O_8)$), potash feldspar $(K(AlSi_3O_8))$, pyrophyllite $(Al_2Si_4O_{10}(OH)_2)$, and natural zeolite. Feng et al. [85] found that it was necessary to calcine albite at 1000 °C with NaOH or Na_2CO_3 as activator in order to reach acceptable compressive strength (43–44 MPa at 28 days) in one-part geopolymer. Albite is often found in mine tailings [86,87] and thus could represent a potential low-cost precursor.

Red mud is a by-product from the production of alumina from bauxite by the Bayer's process. It is characterized by high pH (10–12.5), high solids content (15–30%), and varying chemical composition: Fe_2O_3 20–60%; Al_2O_3 10–30%; SiO_2 2–20%; Na_2O 2–10%; CaO 2–8%; TiO_2 and other oxides up to 28% [88]. Red mud is generated at a rate of approximately 1–2.5 tons per ton of alumina or, 55–65% of the bauxite processed [88,89]. The total worldwide annual production of red mud is approximately 120 million tons, with an existing reservoir of over 2.7 billion tons [90]. There have been efforts to utilize red mud in OPC production [91,92] as well as in alkali-activated binders [93–95]. In one-part geopolymers, red mud has been used as an aluminosilicate source after thermal activation with NaOH at 800 °C [74,96,97]. Additionally, red mud has been used as an alkali source for fly ash in one-part mixes [98]. While it was possible to replace NaOH by red mud as a solid activator, the obtained compressive strengths were poor (< 4 MPa, 28 d) due to the relatively high unburned carbon particle content (loss on ignition > 6%) in the employed fly ash [98].

Van Deventer et al. [64] stated that aluminosilicate material in one-part geopolymer mixes can include any naturally occurring aluminosilicates (such as shales, clays, feldspars, plagioclase, feldspathic materials, zeolite, pyroxene, and amphibole) or industrial by-products (such as mine tailings, fly ashes, slags, red mud, waste glass, and discards from coal or ore mining). Furthermore, they proposed that some of the aluminosilicate could be added before calcination and the rest (0–97% w/w of the dry mix) after calcination [64]. The most suitable aluminosilicate source for one-part mixes depends on the local availability of materials.

2.2. Additional amorphous silica and alumina sources

Reactive amorphous silica and alumina are usually provided in part by aluminosilicate precursors (such as fly ash, metakaolin, or slags) but they are frequently supplemented by an aqueous solution of sodium or potassium silicate (i.e., water glass) or aluminate in conventional two-part geopolymers. One-part mixes, however, use various solid amorphous silica sources (Table 1). In terms of additional reactive alumina, solid sodium aluminate is the most common material and it can also act as an activator (see Section 2.3).

Rice hull and husk ashes are residues from the combustion of agricultural wastes and they can have a high reactive silica content (≥ 90 weight-%) [99,100]. Global rice husk output has been estimated to be 80 Mt., with China as the largest contributor [101]. Rice hull and husk ashes have been activated by solid sodium aluminate in one-part geopolymer mixes [102,103]. It was found that after just one day of curing, compressive strength was relatively high (30 MPa) and that the rice husk ash had reacted almost completely [103]. While these ashes sometimes contain a high amount of unburned carbon [104], it was shown that low-quality rice husk ashes (loss on ignition of 40%) could be successfully utilized in one-part geopolymers [102]. Another potential agricultural residue sources are maize stalk and cob ashes which have high pH (approximately 13–14 in water using solid/water = 3.2), potassium content (approx. 30 weight-% as K_2O), and silica amount (approx. 20 weight-% as SiO_2): they have been used as solid activators of metakaolin or fayalite slag [80,105].

Silica residue from chlorosilane production has been used in one-part geopolymers in combination with sodium aluminate [106,107]. Approximately 5000–10,000 tons per year of silica residue is produced at a typical chlorosilane plant during flue-gas scrubbing and neutralization of wastewater [108]. Silica content of the silica residue is approximately 84–89 weight-%, but the chloride content of the material can be relatively high (0.86–1.36 weight-%) [106–108].

Geothermal silica, another potential source of amorphous silica, commonly forms as precipitate in pipes and on other surfaces at geothermal power plants where water supersaturated with silicic acid (i.e., geothermal fluid) is utilized in electricity generation [109]. In a study using geothermal silica in one-part geopolymers, it was first washed with distilled water at 80 °C (liquid to solid ratio of 10), filtered, washed again, dried, milled, and finally mixed with solid sodium aluminate [110]. The purpose of this rather complicated pretreatment was to remove salts; the resulting material had a silica content of 96% [110].

Silica fume (or microsilica) which is a by-product of silicon and ferrosilicon alloy production, has also been used in one-part geopolymers [97,106,107]. In one study, addition of up to 20–25% silica fume had a clearly positive effect on the compressive strength of a red mud–based one-part geopolymer [97]. Silica fume consists of finely dispersed (95% of particles finer than 1 μm) amorphous SiO_2 (> 92 weight-%), the surface of which is covered by reactive silanol groups [111].

In addition to the aforementioned industrial by-products and waste materials, synthetic silica chemicals such as solid sodium metasilicate (Na_2SiO_3) can also be used to provide additional silica (see Table 1) in one-part geopolymers. Technically, all of the silica sources described here could be used but the most industrially feasible option depends on local availability.

2.3. Alkali sources (activators)

According to Provis [7], the alkali source (i.e., activator) in a one-part geopolymer mix can be any substance that provides alkali cations, raises the pH of the reaction mixture, and facilitates dissolution. Activators employed in one-part geopolymer mixes include solid NaOH, Na_2SiO_3, $Na_2SiO_3 \cdot 5H_2O$, Na_2CO_3, $NaAlO_2$, $CaSO_4$, Na_2SO_4, KOH, red mud, and maize stalk and cob ash (Table 1). The modulus (SiO_2/Na_2O molar ratio) of commercial solid sodium silicates has varied between 0.93 and 3.32 in the published one-part geopolymer studies [66,67,69]. Anhydrous sodium metasilicate (Na_2SiO_3, modulus 0.93) resulted in higher compressive strength and better workability than sodium silicate pentahydrate ($Na_2SiO_3 \cdot 5H_2O$, modulus 1.00) or unspecified hydrous sodium silicate ($Na_2SiO_3 \cdot nH_2O$, modulus 2.06) when activating fly ash and blast furnace slag with or without hydrated lime in one-part

geopolymer [65]. Choo et al. [98] innovatively used red mud as a source of NaOH. In addition, CaO, MgO, dolomite ($CaMg(CO_3)_2$), and $Ca(OH)_2$ have been used in one-part geopolymer mixes (Table 1). These materials supply alkaline earth cations instead of alkali cations, which facilitates the formation of different binding phases compared to systems with a low calcium content [112]. Kim et al. [113] found CaO powder to be a more efficient activator for blast furnace slag than Ca$(OH)_2$ powder although CaO generates higher heat of hydration.

Many of these activators, however, have disadvantages. For example, solid NaOH is corrosive, hygroscopic, and forms sodium carbonate when exposed to CO_2. Currently, NaOH is produced at a rate of approximately 60 Mt per year via the chlor-alkali process and upscaling this production is not straightforward because chlorine (Cl_2), which has a limited world-market, is produced as a side-product [8,46]. The production of synthetic alkali silicates is characterized by a high energy use: the process alternatives are direct fusion of sand and sodium carbonate at 850–1088 °C or evaporation of metasilicate solution [114]. Therefore, replacing synthetic alkali silicates with other alkali and silica sources is important. Na_2CO_3, one alternative activator, is available as a geological resource in the form of trona (Na_2CO_3·$NaHCO_3$·$2H_2O$). Na_2CO_3 has been found to be at least as effective as NaOH in thermal activation of kaolin, bentonite, or albite [85,115,116]. However, Na_2CO_3 and dolomite release CO_2 if used with high-temperature calcination of aluminosilicate precursors. Kovtun et al. [117] published very promising results with solid Na_2CO_3 and slaked lime as the activators (without calcination) for blast furnace slag and silica fume: they obtained up to approximately 50 and 85 MPa compressive strength (28 d) at 25 and 85 °C curing temperatures, respectively. $NaAlO_2$ is an interesting alkali source as it also provides supplementary reactive aluminum. $NaAlO_2$ is produced commercially by dissolving aluminum hydroxides (such as gibbsite from the Bayer process) in sodium hydroxide solution, digesting bauxite at elevated temperature (150–230 °C) and pressure ($\geq$ 5 bar), or processing gibbsite in rotary sintering kilns (at 1000 °C) [118].

2.4. Admixtures

High range water reducers (superplasticizers) are often used to improve workability, rheology, and mechanical properties in OPC concrete or mortar. Commonly used superplasticizers include lignosulfonates, naphthalene, melamine-based compounds, and modified polycarboxylates. However, many of the superplasticizers work poorly with geopolymer pastes due to the extremely alkaline conditions and subsequent degradation of superplasticizer as summarized by Nematollahi et al. [55]. However, naphthalene [119–123] and modified polycarboxylates [124–126] have shown promising performance in some studies involving two-part geopolymers. Ye et al. [97] used sodium lignosulfonate (0.5 weight-%) in a one-part red mud–based geopolymer mixture and were able to reduce the water/solid ratio from 0.55 to 0.45, which improved compressive strength. Yang et al. [127] used a polycarboxylic-based water reducer (0.65 weight-%) in one-part alkali-activated blast furnace slag.

One-part geopolymer pastes frequently set too rapidly due to heat generated from the dissolution of solid activators such as sodium hydroxide [128]. Consequently, using set retarding admixtures to prolong setting time would be beneficial. Nano-ZnO can retard the setting of alkali-activated slags by affecting the nucleation of the C-(N)-A-S-H gel via binding Ca^{2+} into a calcium zincate phase [129]. However, it has not been studied with one-part mixes. Sodium phosphate [130] and phosphoric acid [131] have also been found to retard hydration reaction of (two-part) alkali-activated blast furnace slag. The mechanism of phosphate-based retardants has been suggested to be the formation of sparingly soluble but temporary $Ca(H_2PO_4)_2$ and $CaHPO_4$ phases [132]. Other potential retardants (again, not studied for one-part mixes) include sucrose [133], sodium chloride and other inorganic salts (which are accelerators at low dosages and retarders at high for alkali-activated

blast furnace slag) [134], or malic acid [134]. Wang et al. [66] used unspecified phosphate-based retardant (optimum dose 7%), but they did not report results without retardant. Another potential retardant for one-part geopolymers is tartaric acid [64].

Seeded nucleation, by adding 0.5 weight-% nano-sized Al_2O_3, ZnO, or ZrO_2 to one-part geopolymer, affected positively compressive strength [135]. More specifically, nucleation prevented alumina from sorbing on the surface of silica particles; controlled the silica release rate; and led to the formation of a silica-rich phase in the early stage of geopolymerization [135]. This kind of nucleation phenomena could, however, also occur without the addition of oxides if the raw materials contain suitable impurities.

2.5. Fiber reinforcements

Polyethylene (PE) and polyvinyl alcohol (PVA) fiber reinforcement of one-part geopolymers prepared using fly ash and blast furnace slag or lime has been studied by Nematollahi et al. [67,68,136–139] in several papers. Typically, the amount of fiber added in their studies was 2 vol-%. The compressive strength of these composites is summarized in Table 1; other properties, such as tensile performance, interaction of fibers with geopolymer matrix, and fracture properties, are beyond the scope of this review. However, these studies demonstrate that one-part geopolymer matrixes can be successfully used to develop composites similar to two-part geopolymers.

2.6. One-part geopolymers with OPC (hybrid and alkaline blended cements)

Binders containing OPC, aluminosilicate precursor, and which are alkali activated are referred to as hybrid or blended alkaline cements [140–144]. The cementitious gels in this type of binders consist of a complex mixture of (C,N)-A-S-H and/or N-(C)-A-S-H [140]. However, in this section, only those blended cement studies are considered in which the alkali activator is in a solid form.

Garcia-Lodeiro et al. [145] produced hybrid cements incorporating OPC (60%), and fly ash and bottom ash (40%) provided by municipal solid waste incinerator. A mix of solid $CaSO_4$ and Na_2SO_4 in different percentages was used as an activator. The liquid to solid ratio was kept constant (0.5). They measured the compressive strength and flexural strength higher than 32 MPa and 5 MPa, respectively, after 28 days curing at temperature 21 °C and relative humidity 99%. The results depicted that the developed material had acceptable strengths for using in practical applications [145].

Fernández-Jiménez et al. [146] developed hybrid cements incorporating a low OPC content (20–30%) and fly ash (70–80%). The mixtures were activated either by the addition of water, 8 M sodium hydroxide solution, or solid sodium silicate and water. The liquid to binder ratios varied in the rage of 0.4–0.45 regarding the mixture ingredients. The compressive strength varied in the range of 20–40 MPa (at 28 d, curing temperature 22 °C, and relative humidity 99%), in which the minimum and maximum compressive strengths were detected in the mixtures that used water and solid sodium silicate with water, respectively. The binder with the maximum strength contained calcite and ettringite of which the latter yielded a denser structure partially explaining the higher strength.

García-Lodeiro et al. [147] reported the results of two different hybrid cement systems comprised of OPC (20%) blast furnace slag (40%), dehydroxylated (2 h at 750 °C) kaolin or bentonite (40%), and solid sodium carbonate (5%) as an activator. The two cements had similar CaO/SiO_2 ratios but very different SiO_2/Al_2O_3 ratios (almost 4 for the bentonite mix and 2 for the metakaolin mix). The compressive strengths of 32 MPa and 15 MPa (2 d, curing temperature 22 °C, and relative humidity 99%) were recorded for the systems containing bentonite and metakaolin, respectively. The main differences in the results obtained is likely caused by the different water content (the L/S ratio was 0.3 and 0.5 for bentonite and metakaolin mixture, respectively).

The differences in the water demand affected the hydration products and strength development in these systems. Regarding the microstructure studies, it was revealed that C-S-H and C-A-S-H gels prevail in high calcium and silicon systems, whereas in silicon- and aluminum-rich systems (N,C)-A-S-H and C-A-S-H gels predominated. However, the early stage compressive strengths indicated a very promising performance from the application point of view.

In a study by Abdollahnejad et al. [148,149], one-part hybrid cement mixtures were comprised of OPC, fly ash, kaolin, solid sodium hydroxide, calcium hydroxide, water, and a lignosulfonate-based superplasticizer. Lower reactivity was reported for the mix compositions using sodium hydroxide than for the mix compositions using potassium hydroxide. Efflorescence, which is common in two-part geopolymer mixes, was reduced in one-part geopolymers, possibly due to the smaller amount of sodium or potassium used. In addition, during the hydration process, some sodium and potassium may have been replaced by calcium. The maximum compressive strength reported in this study was around 27 MPa (at 28 d). Microscopic images also confirmed that a dense interfacial transition zone was formed between aggregates and matrix. Finally, they showed that further analyses of life-cycle and cost efficiencies of these blended cements need to be carried out to confirm their eco-efficiency [150].

Moreover, Van Deventer et al. [64] published a patent describing several one-part geopolymer mix compositions containing OPC (3–30%). The highest compressive strengths (35 MPa) at the age of 28 days were obtained in mix compositions containing 30% OPC. In addition, the measured setting time was 2 h at 20 °C and 1 h 15 min at 35 °C, which provides an appropriate time in real construction applications.

2.7. Raw material calcination

Calcination of certain aluminosilicate raw materials is necessary in order to render them sufficiently reactive. For example, for dehydroxylation of the commonly used kaolin into metakaolin (at approximately 600 °C), the main phenomena are the transformation of octahedral Al into tetrahedral Al, structure amorphization, and retaining a 1:1 layer type [151]. With one-part geopolymer mixes, an alkali source, such as NaOH, is commonly added to the calcination phase and the process is referred to as alkali fusion. The addition of sodium to the aluminosilicate increases the number of non-bridging oxygens [85]. It is notable that in some cases, calcination without the alkali addition was not sufficient to alter the structure into reactive form (e.g., with albite up to 1000 °C) [85]. In addition to calcination, mechanochemical treatment (e.g., high intensity ultra-fine grinding) can also be utilized in activation [75].

Bentonite [116,152], kaolin [115,153], halloysite [154], albite [85], and red mud [74,96] are examples of aluminosilicates used in one-part geopolymers that have been calcined (at 550–1100 °C) with the alkali fusion method (see Table 1). The optimum calcination temperature is dependent on the material used: for instance, in the case of bentonite activated by dolomite or Na_2CO_3, calcination at 1100 °C was better than calcination at 1200 °C [152]. Interestingly, quartz, which is commonly present as an impurity in bentonite, became a reactive glassy phase at 1000 °C with 20% NaOH present [152]. However, increasing the calcination temperature can also increase crystallization of aluminosilicate phases, which has a negative effect on geopolymerization due to the unreactivity of such phases. Peng et al. [116] concluded that Na_2CO_3 is more effective than NaOH in high-temperature alkali fusion of bentonite. Increasing the calcination temperature of kaolin with NaOH or Na_2CO_3 up to 950 °C had a positive impact on the compressive strength [116].

However, calcination of raw materials introduces an additional step to the manufacturing of one-part geopolymers and can increase the carbon footprint. In some cases, the activators that are added during calcination, such as dolomite or Na_2CO_3, also introduce CO_2 emissions.

Numerous studies have demonstrated (see Table 1) that calcination can be avoided with the appropriate selection of aluminosilicate raw materials (such as fly ash and blast furnace slag), although, fly ash and blast furnace slag themselves have been formed in high-temperature processes.

2.8. Curing conditions

One-part geopolymers have been cured at both ambient (approximately 25 °C) and at elevated (40–80 °C) temperatures depending on precursors, mix design, and so on (Table 1). Suwan and Fan [128] pointed out that heat generated as a result of the dissolution of solid activators in one-part geopolymers could have a beneficial effect on the curing. Peng et al. [152] obtained significantly lower 28-day compressive strength for one-part samples cured for three days at 20 °C (< 5 MPa) compared to 80 °C (up to 38.3 MPa). Generally, heat curing accelerates the early strength development of geopolymers [64,78].

In addition to temperature, relative humidity is another important factor to control during curing. It is generally preferable to seal geopolymer concrete since dehydration results in efflorescence, microcracking and a subsequent decrease in compressive strength [155]. The heat typically generated upon addition of water to one-part geopolymers promotes water loss and increases the importance of sealing. According to patent literature [64], the preferable curing occurs at saturated conditions (i.e., relative humidity of 100%) and there is no need to add water during curing. In addition to curing in air, one-part geopolymers have also been cured in water tanks [67].

2.9. Mix designs and molar compositions

Generally, increasing the SiO_2/Al_2O_3 molar ratio increases the compressive strength and elasticity up to a certain ratio [156]. Porosity, in contrast, increases at low SiO_2/Al_2O_3 ratios [157]. Typical two-part geopolymer compositions have a SiO_2/Al_2O_3 molar ratio in the range of approximately 3.3–4.5, but if waste precursors are used, this range might not apply [50]. Reported SiO_2/Al_2O_3 molar ratios in one-part geopolymers range from 0.75 to 6.02 (Table 1).

When the SiO_2/Al_2O_3 ratio of metakaolin-based one-part geopolymer was increased from 1.75 to 2.25, the compressive strength decreased [158]. The opposite trend was observed for fly ash–based one-part geopolymer: SiO_2/Al_2O_3 of 1.8 resulted in a higher compressive strength than SiO_2/Al_2O_3 of 2.25 [69]. Increasing the SiO_2/Al_2O_3 molar ratio from 2 to 6 increased the compressive strength of a microsilica-based one-part geopolymer while it decreased the strength of a one-part geopolymer that used chlorosilane production residue [106]. These examples demonstrate that the trend of compressive strength as a function of the SiO_2/Al_2O_3 ratio is not constant across materials and depends on additional factors.

A higher SiO_2/Al_2O_3 ratio decreased the amount of Si in the geopolymer gel and the geopolymerization reaction rate in a fly ash–based one-part geopolymer [69]. One reason might have been the formation of larger, less reactive silica oligomers instead of small reactive species at high SiO_2/Al_2O_3 ratios [69]. In a metakaolin-based one-part geopolymer, a similar reaction rate decrease was observed as the SiO_2/Al_2O_3 ratio increased [158]. Furthermore, increasing the SiO_2/Al_2O_3 ratio caused silica-rich phases to form and increased the amount of unreacted silica [158]. Interestingly, the dissolution of silica from microsilica or chlorosilane production residue halted when the SiO_2/Al_2O_3 ratio reached approximately 2, whereas rice husk ash reacted almost completely at that ratio [103,107,159].

The amount of alkali in the mix must be sufficiently high to cause dissolution of the aluminosilicate (indicated by the molar ratio of M_2O/SiO_2 ratio, where M = Na or K). However, too high alkali concentration causes, for instance, efflorescence (indicated by the M_2O/Al_2O_3 molar ratio, which should optimally be near 1). By increasing the concentration of alkali, the dissolution rate of solid aluminosilicate

increases and the difference between the Al and Si dissolution rates becomes smaller [69]. Peng et al. [152] found that the compressive strength of a one-part geopolymer was noticeably smaller if no alkali (Na_2CO_3) was added to the calcination of bentonite. The Na_2O/SiO_2 and Na_2O/Al_2O_3 molar ratios have varied within 0.16–2.92 and 0.98–1.50, respectively, in the published one-part alkali activated material studies (Table 1). In those cases where alkaline earth metals were present, the $(Ca + Mg)O/SiO_2$ ratio has been 0.02–2.1 (Table 1). A higher CaO/SiO_2 or MgO/SiO_2 molar ratio resulted in higher compressive strength when bentonite-based geopolymer was prepared [152].

Water demand (i.e., the amount of water needed to form a paste of standard consistency) is affected by several factors, including particle size and shape distribution and specific surface area [160]. Furthermore, the heat generated from one-part geopolymer reactions due to dissolution of the solid activators can cause significant loss of water [128]. Water-to-binder ratios (i.e., liquid-to-solid, L/S, where the binder is the amount of solid activators and precursors) of 0.2–0.75 have been studied with one-part geopolymers (Table 1). As a general trend, less water leads to a higher compressive strength [69,127]. As discussed earlier, Ye et al. [97] were able to reduce the amount of water necessary by adding sodium lignosulfonate (from L/S 0.55 to 0.45). With one-part geopolymer containing fly ash and solid sodium silicate, higher water content was seen to result in faster formation of Al-rich gel, substitution of Si, and formation of Si-rich gel [69]. Another way of expressing the amount of water is H_2O/Na_2O or H_2O/Al_2O_3 molar ratios, which have been 7–19 and 7–28, respectively, in one-part geopolymer studies (Table 1). Increasing the H_2O/Na_2O ratio from 14 to 17 decreased the geopolymerization reaction rate and sodium silicate dissolution [158].

3. Properties of one-part geopolymers

The unconfined compressive strength (UCS) of one-part geopolymers varies widely, as can be seen from Table 1. The highest 28-day compressive strength has been up to 80 MPa, whereas also very low values (< 5 MPa) have occasionally been reported. In some cases, the compressive strength of one-part geopolymers has decreased as curing time increases, which has been explained by depolymerization, carbonation, or efflorescence [96,97]. Compressive strength of one-part geopolymers in comparison to similar composition two-part geopolymers has been lower in some cases [65,136], but higher values have also been reported [115]. Yang et al. [62] found a directly proportional correlation with the alkali quality coefficient (Eq. 2, where oxides are given as weight in grams in the binder and B is the total weight of the binder in grams) and rate of flow loss and compressive strength in a one-part geopolymer mix.

$$Q_A = \frac{\left(\frac{Na_2O}{(SiO_2)^2} \times Al_2O_3 \times CaO\right)}{B} \quad (2)$$

The required compressive strengths for different classes of concrete are specified, for example, in the European standard EN 206–1 [164]: the required 28 d values for normal and heavyweight concrete are 8–100 and 10–115 MPa for a cylindrical sample (diameter 150 mm, height 300 mm) and a cubic sample (side length 150 mm), respectively. For lightweight concrete, the similar requirements are 8–80 and 9–88 MPa [164]. These compressive strength classes specify the characteristic compressive strength required in different applications: the lowest strength concretes are used in different non-structural applications (a review by Hooton [21] summarizes several other standards specifying compressive strength requirements). Consequently, one-part geopolymers appear promising in meeting these requirements. However, the comparison of the specified values to those in Table 1 is not straightforward due to, for instance, different binders, aggregates, curing conditions, and measurement set-up (specimen dimensions,

compression loading rate, etc.) employed in one-part geopolymer studies.

Yang et al. [127] also characterized lightweight mortars based on one-part alkali-activated blast furnace slag for splitting tensile strength, flexural strength (i.e., modulus of rupture), and modulus of elasticity (i.e., Young's modulus), which all decreased as the portion of lightweight aggregate to sand was increased. Furthermore, the curves of stress-strain under compressive loading indicated ductile behavior when the amount of lightweight aggregates was > 50% in relation to sand [127]. Shrinkage strain was approximately 100 μm up to 30 days but increased substantially (up to 1300 μm) after that with a higher amount of lightweight aggregates increasing shrinkage [127]. Otherwise, the published information on the shrinkage of one-part geopolymers is scarce.

Workability of fresh one-part geopolymer paste according to the slump test (i.e., spread-flow test, ASTM standard C1437 [165]) has been reported to be 3.52–3.69 as the relative slump value [67,68]. It was noticed that blast furnace slag without gypsum resulted in a higher relative slump value than slag with gypsum in one-part geopolymers [67]. Fly ash and blast furnace slag one-part geopolymers were reported to have thixotropic behavior, that is, good workability during mixing and vibration [68,136]. Increasing the amount of sodium silicate activator in fly ash– and blast furnace slag–based one-part geopolymers resulted in a significant decrease in workability [65]. On the other hand, increasing water content improved workability (but decreased compressive strength) [65]. Workability of one-part geopolymers in comparison to similar composition two-part geopolymers was approximately 35% lower [65].

Initial and final setting times of one-part geopolymers have varied between 23–150 and 69–230 min, respectively [66,75,163]. The initial setting time should be > 45–75 min depending on the strength class of cement according to the standard EN 197–1 [166]. Setting time requirements according to several other standards have been summarized by Hooton [21]. Nematollahi et al. [65] reported that the setting time increased when the blast furnace slag content in a one-part geopolymer mixture containing fly ash was increased. Setting time increased also when the amount of solid calcium carbonate amount decreased, solid sodium hydroxide was kept constant, and the amount of blast furnace slag was increased [163]. Yang et al. [62] observed that alkali-activated blast furnace slag hardened more slowly when using solid activator instead of solution. It should be noted that there are no publications available about the effect of retardants on the setting time of one-part geopolymers.

The heat resistance of one-part geopolymers, like two-part geopolymers [12–14], is claimed to be better than OPC-based concrete [64]. One-part geopolymers based on microsilica and chlorosilane production residue (activated with sodium aluminate) could withstand temperatures up to 700 °C and, in fact, their compressive strength typically increased [106]. Furthermore, shrinkage behavior seemed to correlate with strength development: up to approximately 750 °C, shrinkage of 1–3% was observed, while at 800–900 °C, samples shrunk approximately 10–20%. Shrinking was possibly due to viscous sintering or crystallization behavior. Those samples with increased strength at 800 °C swelled at 800–900 °C, which was explained by the presence and reactions of residual microsilica [106]. Those samples that swelled contained either completely amorphous reaction products or nepheline at temperature above 750 °C as identified by XRD [106].

Water absorption is dependent on the pore structure of material. The water absorption values of one-part alkali-activated blast furnace slag were 7–16%, and these values decreased as curing time increased [163]. This was likely due to the formation of hydration products and thus decreasing porosity. Furthermore, high water to cement ratio or calcium carbonate amount increased water absorption [163]. However, the water resistance of a one-part geopolymer prepared with bentonite has been an issue, as shown by the softening coefficient (i.e., compressive strength at 4 days, of which 1 day of curing under water

divided by the 3-day air-cured compressive strength) [116,152]. Otherwise, no such problems have been documented; in fact, Gluth et al. [108], for example, demonstrated that their one-part geopolymer had good water resistance.

Some authors have measured the pH of fresh one-part geopolymer pastes to evaluate reactivity: for example, Matalkah et al. [75] found that mechanochemically processed fly ash, quick lime, sodium hydroxide, and magnesium oxide resulted in a less intense and slower increase of pH compared to separately milled raw materials. One-part geopolymer pastes prepared of residue from chlorosilane production or silica fume (activated by sodium aluminate) had pH values of 13.4–14.7 [106]. The relatively high pH values were proposed to be related to the formation of hydrosodalite and transformation of zeolite A into hydrosodalite in their one-part geopolymers [106]. Red mud– and silica fume–based one-part geopolymers, on the other, had pH values of 11.2–13.2 when crushed sample (particles < 2.38 mm) was leached with deionized water [97].

4. Geopolymerization mechanisms and binding phases

It has been suggested that the following four steps occur after adding water to one-part geopolymer mixtures: (1) ion exchange, (2) hydrolysis, (3) network breakdown, and (4) release of Si and Al [75]. After these steps, the formation of one-part geopolymers most likely follows the same main steps as two-part geopolymers: speciation, gelation, reorganization, and polymerization [37]. The mechanisms of (two-part) alkali activation of aluminosilicates have been discussed in numerous papers [112,167–170].

An important difference between two and one-part geopolymers is the release rate and availability of Si and Al. In particular, the solid silica sources employed in one-part mixes react more slowly than soluble silicate solutions [171]. Hajimohammadi et al. [171] varied the availability of silica by mixing solid geothermal silica and a sodium silicate solution, with solid sodium aluminate as a source of alumina. Their main observations were that the low silica availability (i.e., using solid geothermal silica) led to the formation of a high-silica-content crystalline phase, analcime ($NaAlSi_2O_6 \cdot H_2O$); nucleation took place near the unreacted silica particles; and the structure was more heterogeneous. Another study by Hajimohammadi et al. [161] addressed the effects of alumina release rate: they prepared a one-part geopolymer mix from fly ash and geothermal silica with either sodium aluminate (fast release) or amorphous alumina (slow release) as a supplementary alumina source. The main observations were that faster alumina release lead to faster formation of Al-rich gel followed by Si-rich gel; a more homogenous geopolymer gel structure; and a high initial concentration of dissolved alumina that prevented silica dissolution, resulting in high early strength but lower subsequent strength development. As summarized by Duxson and Provis [41], the availability of Al is one of the most important parameters affecting properties of geopolymers such as final strength, setting, flexural strength, acid resistance, microstructure, and strength development.

Matalkah et al. [75] observed three exothermic reactions (by isothermal calorimetry) after addition of water to fly ash–based one-part mixture and assigned them as follows: (1) dissolution of NaOH and hydration of CaO (immediately after water addition); (2) attack of OH^- on the Si—O and Al—O bonds (after approximately 130 min); and (3) polymerization (after approximately 170 min). Wang et al. [66] found only two distinctive peaks with calorimetry when observing one-part mixture reactions, and they suggested that both were associated with the dissolution of raw materials: Si—O—Si and Al—O—Al bond breaking; release of Ca, Si, and Al; and the formation of gel covering unreacted slag particles. Kim et al. [113] found two peaks by calorimetry: the first was attributed to wetting and dissolution of CaO, $Ca(OH)_2$, and blast furnace slag while the second was not explained. X-ray photoelectron spectroscopy (XPS) studies have revealed that Si and Al (both tetrahedral) in albite become more ionized and disordered after calcination

with NaOH or Na_2CO_3, and thus, addition of water would cause repolymerization [85]. Furthermore, magic-angle spinning (MAS) NMR studies of ^{29}Si have revealed that in the blast furnace slag– and fly ash–based geopolymer, the Si environment evolved from a combination of Q^0, Q^1, and Q^2 in the unreacted blast furnace slag to Q^2 and $Q^2(1Al)$ in the cured one-part mixture similar to two-part alkali activated materials [66]. In a separate study, the proportion of Q^4 and $Q^0(4Al)$ Si environments has been found to increase when the SiO_2/Al_2O_3 ratio increases and Q^3 and $Q^3(1Al)$ to decrease [107]. This was explained by the presence of unreacted microsilica at higher SiO_2/Al_2O_3 ratios [107].

One major factor affecting the composition of binding phases of alkali-activated (two-part) materials is the calcium content: N-A-S-H, C-(N)-A-S-H, and C-A-S-H gels are formed in low-, intermediate-, and high-calcium systems, respectively [142,172–177]. In one-part geopolymers, after the addition of water and curing, similar N-A-S-(H) gels have been proposed to be the binding phases [96,103]. However, these studies concluded the presence of N-A-S-(H) gel indirectly via XRD and FTIR analyses (N-A-S-(H) gel has disordered structure and thus it is not directly observable by XRD). No differences were found in the reaction products of one and two-part fly-ash geopolymers when prepared with similar mixing ratios [128]. In high-calcium systems, the main binding phase has been observed to be crystalline (i.e., observable with XRD) C-S-H [63,97,113]. It is likely that also C-A-S-H gel is produced in the high-calcium one-part systems. In addition, several studies have identified different zeolites and unspecified amorphous phases as the hydration products [96,103,106,108,116,152]. Hajimohammadi and van Deventer [69] suggested, however, that one-part geopolymers might have a lower tendency for zeolite formation than two-part geopolymers. Matalkah et al. [75] detected that portlandite was not formed when coal fly ash (with a relatively high CaO content of 14.3%) was milled together with alkali activator (quick lime, NaOH, and MgO) before adding water, whereas separate milling produced portlandite. According to García-Lodeiro et al. [143], portlandite formation is promoted by a lower alkalinity: for instance, by the use of sodium carbonates or sulfates instead of sodium hydroxide. Examples of other observed crystalline secondary hydration products include cancrinite, akermanite, hydrous gehlenite, sodium aluminum silicate, wairakite-Ca, magnesium aluminum hydroxide, katoite, brucite, gibbsite, stilpnomelane, calcium hydroxide, katoite, and tobermorite depending on the precursors, activators, reactions conditions and so on [97,152]. As the number of studies systematically comparing the reaction products of one and two-part alkali-activated materials is still scarce, it is yet difficult to assess if there are clear differences in the nature of (secondary) hydration products.

The reactive minor phases in one-part geopolymers consist of aluminum, magnesium and iron minerals able to hydrate (i.e., react with water) as discussed below. Peng et al. [116] found that calcium ferrite and amorphous phases were reactive when water was added to dry mix prepared of bentonite calcined with Na_2CO_3 or NaOH. In another study by Peng et al. [152], C_3A (tricalcium aluminate, $3CaO \cdot Al_2O_3$), C_2S (belite, $2CaO \cdot SiO_2$), and MgO were formed after calcination of bentonite with dolomite and Na_2CO_3. Ke et al. [96] identified C_3A, C_2S, and CAS_2 phases from red mud that was thermally treated with NaOH. These are the same phases that hydrate in calcium aluminate cement to provide strength [38]. Red mud reacted with NaOH at 800 °C contained calcium silicate and sodium aluminosilicate, which hydrated when in contact with water [97].

5. Environmental impact and costs of one-part geopolymers

The environmental impact of OPC concretes and geopolymers is typically evaluated based on a life-cycle assessment analysis (LCA). This "cradle to grave" approach is a normalized method [178] used by industry and academics. The LCA should consider all the possible environmental impacts of a product, from acquisition of raw materials to final disposal. For geopolymers, the net CO_2 savings and global

Table 2

Environmental impact of various geopolymer mixtures. The environmental impact of geopolymer concrete compared to OPC concrete has been reported in various ways in the studies and are not always directly comparable.

One-part/two-part	Solid binder precursors	Added liquid	Environmental impact, % of equivalent OPC concrete	Ref.
Two-part	Fly ash, BFS, metakaolin, NaOH	$(Na_2SiO_2)_nO$	− 20–20%[a]	[180]
Two-part	Coal fly ash	NaOH, $(Na_2SiO_2)_nO$	10%[b]	[181]
Two-part	Coal fly ash, lime, silica fume	Na-Alu (Bayer liquid)	94%[c]	[182]
Two-part	Fly ash, BFS	NaOH, $(Na_2SiO_2)_nO$	74–44%[d]	[183]
Two-part	FCC[e]	Rice husk ash + NaOH	67%[f]	[184]
Two-part	Recycled clay soil, BFS	NaOH, $(Na_2SiO_2)_nO$	4%[g]	[185]
Two-part	Metakaolin, bentonite, silica fume	NaOH, $(Na_2SiO_2)_nO$	40%[h]	[186]
Two-part	BFS, fly ash	NaOH, $(Na_2SiO_2)_nO$	70%	[187]
One-part	OPC, fly ash, kaolin, $Ca(OH)_2$, NaOH	Water, H_2O_2	72%[j]	[188]
One-part	Fly ash, "typical" BFS, "gypsum-free" BFS, $(Na_2SiO_2)_nO$	Water	76%[i]	[67]
One-part	BFS, fly ash, metakaolin, $(Na_2SiO_2)_nO$, NaOH	Water	75–55%[k]	[189]
One-part	BFS, fly ash, $Ca(OH)_2$, $Mg(NO_3)_2$, $(Na_2SiO_2)_nO$	Water (pre-formed foam)	93–85%[l]	[190]
One-part	Fly ash, BFS, thermally activated potash feldspar, NaOH	Water	96%[m], 78%[n]	[191]
One-part/two-part	Fayalite slag or maize cob ash and fayalite slag	Water or NaOH and $(Na_2SiO_2)_nO$	83–53% or 77–55%[o]	[80]
One-part (hybrid cement)	Fly ash, OPC	Water	30%[p]	[192]

BFS = blast furnace slag; OPC = ordinary Portland cement.

[a] Comparison with 30% blended cement, economic allocation.
[b] GWP reduction reported as CO_2-e emissions.
[c] Reported as embodied energy reduction compared to OPC.
[d] Reported as greenhouse gas emissions.
[e] Spent fluid catalytic cracking catalyst.
[f] CO_2 emissions calculated according to the Intergovernmental Panel on Climate Change Guidelines for National Greenhouse Gas Inventories. Geopolymer and OPC mortars were compared.
[g] 19 m^2 geopolymer and OPC paving blocks were compared. GWP reduction percentage calculated by the authors.
[h] OPC and geopolymer binders were compared.
[i] Strain-hardening geopolymer and OPC composites were compared.
[j] Foamed (with 3.5% H_2O_2) geopolymer and OPC thermal insulation mortars were compared.
[k] Reported as carbon footprint.
[l] Foamed geopolymer and OPC thermal insulation concretes were compared.
[m] No GWP allocation on BFS or FA.
[n] GWP allocation for BFS and FA.
[o] Reported as single score environmental impact with an average transport distance of 100 km.
[p] Hybrid cement and OPC pastes were compared. Mix design was retrieved from [142].

warming potential (GWP) is typically evaluated and compared with that of OPC concrete. Other environmental impact categories than GWP include abiotic depletion, ozone layer depletion, human toxicity, freshwater ecotoxicity, marine ecotoxicity, terrestrial ecotoxicity, photochemical oxidation, acidification, and eutrophication. Typically, these categories are not as relevant for construction materials as they are, for example, for agricultural industry [179].

In this section, eight environmental impact studies of two-part geopolymers [180−187], five of one-part geopolymers [67,188−191], and two studies considering both [80,192] were analyzed (Table 2). Overall, the published results are somewhat contradictory: the net GWP change of geopolymers has been reported to range from a reduction of 96% [191] to an increase of 20% [180] in comparison to OPC concrete. In the other environmental impact categories (abiotic depletion etc.), geopolymers have a modest positive or negative impact compared to the impact of OPC concrete [192]. Taking an average of the values reported in Table 2, the environmental impact is 24% for one-part geopolymers and 60% for two-part geopolymers of the environmental impact of OPC. Thus, one-part geopolymers are potentially more environmental friendly than two-part geopolymers.

Ouellet-Plamondon and Habert [192] noted that it is difficult to compare OPC concrete and geopolymer mixes directly. The typical LCA functional unit is one cubic meter of OPC concrete or geopolymer with a given compressive strength. However, this may not be the best approach. It was suggested that also the volume of the paste in the concretes and geopolymers should be the same [192]. Ouellet-Plamondon & Habert [192] re-calculated some of the published results with updated parameters and concluded that although the results did

not change drastically, certain mixes appeared more environmental friendly than initially suggested in the papers.

One aspect that stood out in the environmental impact studies of geopolymers is the large environmental footprint (up to 80–90%) of the sodium silicate solution. The environmental impact of sodium silicate is particularly pronounced if it is produced using the Solvay process. However, some studies [192,193] reported that the data related to sodium silicate production [194] is outdated and that the manufacturers would not disclose information on current energy usage and emissions. Most studies used an older version of the Ecoinvent database to calculate the environmental impact of raw materials. A recent update (version 3) of that database shows that excluding water from the sodium silicate production process considerably reduces the environmental impact of sodium silicates and thus also reduces the environmental impact of alkali-activated materials. The environmental impact of sodium hydroxide is also significant, but much less so than that of sodium silicate. Heath et al. [186] reported that GWP per mole is two times lower for sodium hydroxide than for sodium silicate, but as Habert and Ouellet-Plamondon [191] calculated the CO_{2-eq} per kg of the dry component (i.e. without the water) with the updated data, the difference between sodium hydroxide and sodium silicate was only around 10%. An interesting result was presented by Jamieson et al. [182], who used sodium aluminate solution from the Bayer process as an alkali activator. They found that the embodied energy was as low as 0.33 GJ/t for the geopolymer concrete, representing only 6% of the embodied energy of OPC concrete. These calculations emphasize the importance of finding alternative activators and amorphous silica sources, as discussed in Sections 2.2 and 2.3.

Other relevant environmental impact factors that increase GWP of geopolymers are transportation of raw materials and heat curing. McLellan et al. [183] highlighted that the impact of transport emissions is larger for geopolymers than for OPC in Australia due to the long distances over which feedstocks must be transported. Turner and Collins [181] calculated the emissions for curing for 24 h at 50 °C and found that it contributed 12% of the total CO_2-e for the two-part geopolymer concrete. Thus, even though curing temperatures used for geopolymers are low, the emissions they produce are not negligible.

In addition, the costs of one-part geopolymers are highly dependent on calculation method and product type. Habert and Ouellet-Plamondon [191] compared economic allocation of one-part geopolymers and found that it would be possible to achieve an 80% reduction in costs compared with OPC. Abdollahnejad et al. [188] estimated that the most cost-efficient one-part geopolymer thermal insulation material would cost €67/m^3. Yang et al. [190] concluded that the cost of their one-part alkali-activated BFS foamed concrete was slightly higher, by 1.0 to 1.11 times, than that of OPC concrete. Chan et al. [195] estimated the material and manufacturing costs for over 20 years of FA-based geopolymer cement and OPC and concluded that geopolymer would be 18% more expensive.

6. Conclusions

The development of one-part ("just add water") alkali-activated materials is an important step forward in the commercialization of these alternative, low-carbon binders. Conventional, two-part alkali activation (in which a solid precursor is activated with alkali solution) has several disadvantages: the activating solution is viscous, difficult and non-user-friendly to handle, and transporting aqueous solutions is more expensive than transporting dry activators. However, one-part alkali-activated materials could be best suited for in situ casting while their two-part counterparts might find applications in precast work.

The aluminosilicate sources (precursors) and additional silica or alumina sources that are suitable for one-part alkali-activated materials are largely the same as those for conventional alkali-activated materials: class F fly ash, blast furnace slag, kaolin/metakaolin, albite, bentonite, geothermal silica, silica residue from chlorosilane production, silica fume, fayalite slag, and so on. The most commonly used activators in one-part geopolymer studies are synthetic solid sodium hydroxide and sodium silicate. However, these synthetic chemicals, especially synthetic sodium silicate, do not represent a commercially or ecologically optimized solution when used as the sole activators. Consequently, there have been innovative attempts to replace sodium hydroxide with red mud, sodium carbonate, or calcium oxide, for instance, and sodium silicate with silica fume, rice husk ash, or maize cob ash. The selection of the most suitable raw materials depends on local availability. Furthermore, by choosing the silica and alumina sources carefully, it is possible to adjust the release rate of these elements and consequently modify the strength development, for instance.

Ambient temperature mechanochemical activation (e.g., ball-milling) of the aluminosilicate precursor with the solid activator seems to be a promising method for overcoming the need to use high temperature during geopolymer cement preparation. However, comparison in the energy requirements of thermal and mechanochemical activation should be performed. Numerous examples show that by carefully selecting the solid aluminosilicate, alkali, and supplementary silicate and/or aluminum sources, it is possible to prepare one-part mixes that cure readily at ambient conditions without calcination of the raw materials.

The mechanical properties of one-part alkali-activated materials are promising in terms of compressive strength: up to 80 MPa values at the age of 28 days has been reported. However, other properties, such as durability, efflorescence, and shrinkage, are not yet well documented. Existing studies point out that one-part alkali-activated materials can be reinforced with fibers such as polyvinyl alcohol or polyethylene, similar to its two-part equivalents. However, the rapid setting time of one-part alkali-activated materials is a major shortcoming. The pronounced heat release during hydration accelerates this rapid setting. The use and suitability of retarding admixtures is currently poorly documented.

The binding phases of one-part alkali-activated materials have been identified as similar N-A-S-(H) and C-(A)-S-H gels as with two-part materials. In many cases, the formation of zeolites has been documented as well. The curing mechanism of one-part alkali-activated materials has been specifically addressed in few studies. Calorimetric analysis has shown that typically there is an almost instantaneous reaction after adding water (dissolution of solid activators) followed by slower reactions (dissolution of aluminosilicates). One major difference between one- and two-part alkali-activated materials is in the release rate of silica and alumina species.

The environmental impact of one-part alkali-activated materials is lower than that of two-part equivalents or OPC concrete. The mix design plays a decisive role and specifically the amount of activator are the major factors affecting environmental impact. It is also important to choose the right application for specific geopolymer mixes. The production process and raw materials must be sustainable regarding the environment, but also financially.

In conclusion, there are no obstacles prohibiting the development of commercially feasible "just add water" alkali-activated materials. However, more documentation and testing is still required in order to demonstrate the suitability and long-term durability of these binders. In addition, fundamental understanding on the reaction kinetics is lacking.

Acknowledgements

References

[1] R.M. Andrew, Global CO_2 emissions from cement production, Earth Syst. Sci. Data. Discuss. (2017) In review https://doi.org/10.5194/essd-2017-77.

[2] M. Schneider, M. Romer, M. Tschudin, H. Bolio, Sustainable cement production—present and future, Cem. Concr. Res. 41 (2011) 642–650.

[3] The Cement Sustainability Initiative, Cement Industry Energy and CO_2 Performance, "Getting the Numbers Right", World Business Council for Sustainable Development, Conches-Geneva, 2009.

[4] J.S. Damtoft, J. Lukasik, D. Herfort, D. Sorrentino, E.M. Gartner, Sustainable development and climate change initiatives, Cem. Concr. Res. 38 (2008) 115–127.

[5] R.J. Flatt, N. Roussel, C.R. Cheeseman, Concrete: an eco material that needs to be improved, J. Eur. Ceram. Soc. 32 (2012) 2787–2798.

[6] E. Gartner, H. Hirao, A review of alternative approaches to the reduction of CO_2 emissions associated with the manufacture of the binder phase in concrete, Cem. Concr. Res. 78 (2015) 126–142.

[7] J.L. Provis, Introduction and scope, in: J.L. Provis, J.S.J. Van Deventer (Eds.), Alkali Activated Materials, State-of-the-Art Report, RILEM TC 224-AAM, Springer, Dordrecht, 2014, pp. 1–9.

[8] J.L. Provis, Alkali-activated materials, Cem. Concr. Res. (2017) In press, https://doi.org/10.1016/j.cemconres.2017.02.009.

[9] T.A. Aiken, W. Sha, J. Kwasny, M.N. Soutsos, Resistance of geopolymer and Portland cement based systems to silage effluent attack, Cem. Concr. Res. 92 (2017) 56–65.

[10] M. Albitar, M.S. Mohamed Ali, P. Visintin, M. Drechsler, Durability evaluation of geopolymer and conventional concretes, Constr. Build. Mater. 136 (2017) 374–385.

[11] T. Bakharev, Resistance of geopolymer materials to acid attack, Cem. Concr. Res. 35 (2005) 658–670.

[12] D.L.Y. Kong, J.G. Sanjayan, Effect of elevated temperatures on geopolymer paste, mortar and concrete, Cem. Concr. Res. 40 (2010) 334–339.

[13] P.K. Sarker, S. Kelly, Z. Yao, Effect of fire exposure on cracking, spalling and residual strength of fly ash geopolymer concrete, Mater. Des. 63 (2014) 584–592.

[14] P.K. Sarker, S. McBeath, Fire endurance of steel reinforced fly ash geopolymer concrete elements, Constr. Build. Mater. 90 (2015) 91–98.

[15] K. Sagoe-Crentsil, T. Brown, A. Taylor, Drying shrinkage and creep performance of geopolymer concrete, J. Sustain. Cement-Based Mater. 2 (2013) 35–42.

[16] A.M.M. Al Bakri, H. Kamarudin, M. Binhussain, I.K. Nizar, A.R. Rafiza, Y. Zarina, Comparison of geopolymer fly ash and ordinary portland cement to the strength of concrete, Adv. Sci. Lett. 19 (2013) 3592–3595.

[17] Z. Zhang, J.L. Provis, A. Reid, H. Wang, Fly ash-based igeopolymers: the relationship between composition, pore structure and efflorescence, Cem. Concr.

Res. 64 (2014) 30–41.

[18] I. García-Lodeiro, A. Palomo, A. Fernández-Jiménez, Alkali–aggregate reaction in activated fly ash systems, Cem. Concr. Res. 37 (2007) 175–183.

[19] C. Shi, Z. Shi, X. Hu, R. Zhao, L. Chong, A review on alkali-aggregate reactions in alkali-activated mortars/concretes made with alkali-reactive aggregates, Mater. Struct. 48 (2015) 621–628.

[20] A. Mehta, R. Siddique, An overview of geopolymers derived from industrial by-products, Constr. Build. Mater. 127 (2016) 183–198.

[21] D.R. Hooton, Current developments and future needs in standards for cementitious materials, Cem. Concr. Res. 78 (2015) 165–177.

[22] DSTU B V.2.7-181, Alkaline Cements Specifications, National Standard of Ukraine, Kiev, 2009.

[23] J.S.J. van Deventer, J.L. Provis, P. Duxson, Technical and commercial progress in the adoption of geopolymer cement, Miner. Eng. 29 (2012) 89–104.

[24] H. Kühl, Slag cement and process making the same, US Patent 900,939 (1908).

[25] V.D. Glukhovsky, Soil Silicates, Gostroiizdat Publish, Kiev, 1959.

[26] P.V. Krivenko, Synthesis of Cementitious Materials of the Me₂O-MeO-Me₂O₃-SiO₂-H₂O System with Required Properties (D.Sc. (Eng) Thesis), KISI Public, Kiev, 1986.

[27] J. Davidovits, Geopolymers - Inorganic polymeric new materials, J. Therm. Anal. 37 (1991) 1633–1656.

[28] A. Palomo, F.P. Glasser, Chemically-bonded cementitious materials based on metakaolin, Br. Ceram. Trans. J. 91 (1992) 107–112.

[29] F. Pacheco-Torgal, J. Castro-Gomes, S. Jalali, Alkali-activated binders: a review: part 1. Historical background, terminology, reaction mechanisms and hydration products, Constr. Build. Mater. 22 (2008) 1305–1314.

[30] J.L. Provis, S.A. Bernal, Milestones in the analysis of alkali-activated binders, J. Sustain. Cement-Based Mater. 4 (2015) 74–84.

[31] D.M. Roy, Alkali-activated cements Opportunities and challenges, Cem. Concr. Res. 29 (1999) 249–254.

[32] A. Palomo, P. Krivenko, I. García-Lodeiro, E. Kavalerova, O. Maltseva, A. Fernández-Jiménez, A review on alkaline activation: new analytical perspectives, Mater. Constr. 64 (2014) 140–159.

[33] I. Lecomte, C. Henrist, M. Liégeois, F. Maseri, A. Rulmont, R. Cloots, (Micro)-structural comparison between geopolymers, alkali-activated slag cement and Portland cement, J. Eur. Ceram. Soc. 26 (2006) 3789–3797.

[34] P. Duxson, J.L. Provis, G.C. Lukey, F. Separovic, J.S. van Deventer, 29Si NMR study of structural ordering in aluminosilicate geopolymer gels, Langmuir 21 (2005) 3028–3036.

[35] H. Rahier, W. Simons, B. Van Mele, M. Biesemans, Low-temperature synthesized aluminosilicate glasses: part III Influence of the composition of the silicate solution on production, structure and properties, J. Mater. Sci. 32 (1997) 2237–2247.

[36] J.L. Provis, J.S.J. van Deventer, Introduction to geopolymers, in: J.L. Provis, J.S.J. Van Deventer (Eds.), Geopolymers - Structure, Processing, Properties and Industrial Applications, Woodhead Publishing and CRC Press, Cambridge and Boca Raton, 2009, pp. 1–11.

[37] P. Duxson, A. Fernández-Jiménez, J.L. Provis, G.C. Lukey, A. Palomo, J.S.J. Van Deventer, Geopolymer technology: the current state of the art, J. Mater. Sci. 42 (2007) 2917–2933.

[38] J.L. Provis, Geopolymers and other alkali activated materials: why, how, and what? Mater. Struct. 47 (2014) 11–25.

[39] J.L. Provis, P. Duxson, E. Kavalerova, P.V. Krivenko, Z. Pan, F. Puertas, J.S.J. van Deventer, Historical aspects and overview, in: J.L. Provis, J.S.J. van Deventer (Eds.), Alkali-activated materials: State-of-the-Art Report, RILEM TC 224-AAM, Springer, Dordrecht, 2014, pp. 11–58.

[40] J.L. Provis, Activating solution chemistry for geopolymers, in: J.L. Provis, J.S.J. van Deventer (Eds.), Geopolymers: Structure, Processing, Properties and Industrial Applications, Woodhead Publishing and CRC Press, Cambridge and Boca Raton, 2009, pp. 50–71.

[41] P. Duxson, J.L. Provis, Designing precursors for geopolymer cements, J. Am. Ceram. Soc. 91 (2008) 3864–3869.

[42] A. Purdon, The action of alkalis on blast-furnace slag, J. Soc. Chem. Ind. 59 (1940) 191–202.

[43] R.F. Heitzmann, M. Fitzgerald, J.L. Sawyer, Mineral binder and compositions employing the same, Mineral binder and compositions employing the same. US Patent 4,642,137 (1987).

[44] W. Schwarz, A. Lerat, Tectoaluminosilicate cement and a process for its manufacture, Tectoaluminosilicate cement and a process for its manufacture. U.S. Patent 5,372,640 (1994).

[45] J. Davidovits, Method for obtaining a geopolymeric binder allowing to stabilize, solidify and consolidate toxic or waste materials, Method for obtaining a geopolymeric binder allowing to stabilize, solidify and consolidate toxic or waste materials. U.S. Patent 5,349,118 (1994).

[46] J. Davidovits, Geopolymer Chemistry & Applications, 4th ed., Institut Geopolymere, Saint-Quentin, 2015.

[47] A. Palomo, A. Fernández-Jiménez, C. López-Hombrados, J.L. Lleyda, Railway sleepers made of alkali activated fly ash concrete, Rev. Ing. Constr. 22 (2007) 75–80.

[48] T. Glasby, J. Day, R. Genrich, J. Aldred, EFC Geopolymer Concrete Aircraft Pavements at Brisbane West Wellcamp Airport, in: J. Sanjayan (Ed.), Proceedings of the 27th Biennial National Conference of the Concrete Institute of Australia in conjunction with the 69th RILEM Week, Concrete Institute of Australia, Sydney, 2015.

[49] X.Y. Zhuang, L. Chen, S. Komarneni, C.H. Zhou, D.S. Tong, H.M. Yang, W.H. Yu, H. Wang, Fly ash-based geopolymer: clean production, properties and applications, J. Clean. Prod. 125 (2016) 253–267.

[50] D. Khale, R. Chaudhary, Mechanism of geopolymerization and factors influencing

[51] B. Singh, G. Ishwarya, M. Gupta, S.K. Bhattacharyya, Geopolymer concrete: a review of some recent developments, Constr. Build. Mater. 85 (2015) 78–90.

[52] M.B. Mohd Salahuddin, M. Norkhairunnisa, F. Mustapha, A review on thermo-physical evaluation of alkali-activated geopolymers, Ceram. Int. 41 (2015) 4273–4281.

[53] W.K. Part, M. Ramli, C.B. Cheah, An overview on the influence of various factors on the properties of geopolymer concrete derived from industrial by-products, Constr. Build. Mater. 77 (2015) 370–395.

[54] F. Rao, Q. Liu, Geopolymerization and its potential application in mine tailings consolidation: a review, Miner. Process. Extr. Metall. Rev. 36 (2015) 399–409.

[55] B. Nematollahi, J. Sanjayan, Efficacy of available superplasticizers on geopolymers, Res. J. Appl. Sci. Eng. Technol. 7 (2014) 1278–1282.

[56] F.U.A. Shaikh, Review of mechanical properties of short fibre reinforced geopolymer composites, Constr. Build. Mater. 43 (2013) 37–49.

[57] F. Pacheco-Torgal, Y. Ding, S. Miraldo, Z. Abdollahnejad, J.A. Labrincha, Are geopolymers more suitable than Portland cement to produce high volume recycled aggregates HPC? Constr. Build. Mater. 36 (2012) 1048–1052.

[58] E. Obonyo, E. Kamseu, U.C. Melo, C. Leonelli, Advancing the use of secondary inputs in geopolymer binders for sustainable cementitious composites: a review, Sustain. 3 (2011) 410–423.

[59] J.L. Provis, P. Duxson, J.S.J. van Deventer, The role of particle technology in developing sustainable construction materials, Adv. Powder Technol. 21 (2010) 2–7.

[60] B. Majidi, Geopolymer technology, from fundamentals to advanced applications: a review, Mater. Technol. 24 (2009) 79–87.

[61] K. Komnitsas, D. Zaharaki, Geopolymerisation: a review and prospects for the minerals industry, Miner. Eng. 20 (2007) 1261–1277.

[62] K.H. Yang, J.K. Song, A.F. Ashour, E.T. Lee, Properties of cementless mortars activated by sodium silicate, Constr. Build. Mater. 22 (2008) 1981–1989.

[63] K.H. Yang, J.K. Song, Workability loss and compressive strength development of cementless mortars activated by combination of sodium silicate and sodium hydroxide, J. Mater. Civ. Eng. 21 (2009) 119–127.

[64] J.S.J. van Deventer, D. Feng, P. Duxson, Dry mix cement composition, methods and system involving same, US Patent 7,691,198 B2 (2010).

[65] B. Nematollahi, J. Sanjayan, F.U.A. Shaikh, Synthesis of heat and ambient cured one-part geopolymer mixes with different grades of sodium silicate, Ceram. Int. 41 (2015) 5696–5704.

[66] K. Wang, L. Du, X. Lv, Y. He, X. Cui, Preparation of drying powder inorganic polymer cement based on alkali-activated slag technology, Powder Technol. 312 (2017) 204–209.

[67] B. Nematollahi, J. Sanjayan, J. Qiu, E.-. Yang, Micromechanics-based investigation of a sustainable ambient temperature cured one-part strain hardening geopolymer composite, Constr. Build. Mater. 131 (2017) 552–563.

[68] B. Nematollahi, J. Sanjayan, J. Qiu, E.-. Yang, High ductile behavior of a polyethylene fiber-reinforced one-part geopolymer composite: A micromechanics-based investigation, Arch. Civ. Mech. Eng. 17 (2017) 555–563.

[69] A. Hajimohammadi, J.S.J. van Deventer, Characterisation of one-part geopolymer binders made from fly ash, Waste Biom. Valor. 8 (2017) 225–233.

[70] ASTM, C618–15, Standard Specification for Coal Fly Ash and Raw Or Calcined Natural Pozzolan for Use in Concrete, ASTM International, West Conshohocken, 2015.

[71] P. Chindaprasirt, P. De Silva, K. Sagoe-Crentsil, S. Hanjitsuwan, Effect of SiO₂ and Al₂O₃ on the setting and hardening of high calcium fly ash-based geopolymer systems, J. Mater. Sci. 47 (2012) 4876–4883.

[72] U. Rattanasak, K. Pankhet, P. Chindaprasirt, Effect of chemical admixtures on properties of high-calcium fly ash geopolymer, Int. J. Miner. Metall. Mater. 18 (2011) 364.

[73] P. Duxson, Geopolymer precursor design, in: J.L. Provis, J.S.J. Van Deventer (Eds.), Geopolymers: Structures, Processing, Properties and Industrial Applications, Woodhead publishing Ltd and CRC Press, Cambridge and Boca Raton, 2009, pp. 37–49.

[74] N. Ye, Y. Chen, J. Yang, S. Liang, Y. Hu, B. Xiao, Q. Huang, Y. Shi, J. Hu, X. Wu, Co-disposal of MSWI fly ash and Bayer red mud using an one-part geopolymeric system, J. Hazard. Mater. 318 (2016) 70–78.

[75] F. Matalkah, L. Xu, W. Wu, P. Soroushian, Mechanochemical synthesis of one-part alkali aluminosilicate hydraulic cement, Mater. Struct. 50 (2017) 97.

[76] J. Temuujin, R. Williams, A. Van Riessen, Effect of mechanical activation of fly ash on the properties of geopolymer cured at ambient temperature, J. Mater. Process. Technol. 209 (2009) 5276–5280.

[77] J.N. Djobo, A. Elimbi, H.K. Tchakouté, S. Kumar, Mechanical activation of volcanic ash for geopolymer synthesis: effect on reaction kinetics, gel characteristics, physical and mechanical properties, RSC Adv. 6 (2016) 39106–39117.

[78] X. Li, Z. Wang, Z. Jiao, Influence of curing on the strength development of calcium-containing geopolymer mortar, Mater. 6 (2013) 5069–5076.

[79] H. Shi, Characterization and Modification of the Secondary Copper Smelting Slag for Smooth Operation and Slag Valorization (Ph.D. Thesis), KU Leuven, 2017.

[80] A. Peys, L. Arnout, B. Blanpain, H. Rahier, K. van Acker, Y. Pontikes, Mix-design Parameters and Real-life Considerations in the Pursuit of Lower Environmental Impact Inorganic Polymers, Waste Biomass Valoris. (2017) 1–11.

[81] M. Hu, X. Zhu, F. Long, Alkali-activated fly ash-based geopolymers with zeolite or bentonite as additives, Cem. Concr. Compos. 31 (2009) 762–768.

[82] A. Buchwald, M. Hohmann, K. Posern, E. Brendler, The suitability of thermally activated illite/smectite clay as raw material for geopolymer binders, Appl. Clay Sci. 46 (2009) 300–304.

[83] T. Seiffarth, M. Hohmann, K. Posern, C. Kaps, Effect of thermal pre-treatment

conditions of common clays on the performance of clay-based geopolymeric binders, Appl. Clay Sci. 73 (2013) 35–41.

[84] C. Ruiz-Santaquiteria, A. Fernández-Jiménez, J. Skibsted, A. Palomo, Clay reactivity: Production of alkali activated cements, Appl. Clay Sci. 73 (2013) 11–16.

[85] D. Feng, J.L. Provis, J.S.J. van Deventer, Thermal activation of albite for the synthesis of one-part mix geopolymers, J. Am. Ceram. Soc. 95 (2012) 565–572.

[86] S. Jannesar Malakooti, S.Z. Shafaei Tonkaboni, M. Noaparast, F. Doulati Ardejani, R. Naseh, Characterisation of the Sarcheshmeh copper mine tailings, Kerman province, southeast of Iran, Environ. Earth Sci. 71 (2014) 2267–2291.

[87] M. Khorasanipour, Environmental mineralogy of Cu-porphyry mine tailings, a case study of semi-arid climate conditions, sarcheshmeh mine, SE Iran, J. Geochem. Explor. 153 (2015) 40–52.

[88] R.K. Paramguru, P.C. Rath, V.N. Misra, Trends in red mud utilization - a review, Miner. Process. Extr. Metall. Rev. 26 (2005) 1–29.

[89] A.R. Hind, S.K. Bhargava, S.C. Grocott, The surface chemistry of Bayer process solids: a review, Colloids Surf. A Physicochem. Eng. Asp. 146 (1999) 359–374.

[90] Y. Pontikes, G.N. Angelopoulos, Bauxite residue in cement and cementitious applications: Current status and a possible way forward, Resour. Conserv. Recycl. 73 (2013) 53–63.

[91] I. Vangelatos, G.N. Angelopoulos, D. Boufounos, Utilization of ferroalumina as raw material in the production of Ordinary Portland Cement, J. Hazard. Mater. 168 (2009) 473–478.

[92] P.E. Tsakiridis, S. Agatzini-Leonardou, P. Oustadakis, Red mud addition in the raw meal for the production of Portland cement clinker, J. Hazard. Mater. 116 (2004) 103–110.

[93] E. Jamieson, C.S. Kealley, A. Van Riessen, R.D. Hart, Optimising ambient setting Bayer derived fly ash geopolymers, Mater. 9 (2016).

[94] Q. Nie, W. Hu, T. Ai, B. Huang, X. Shu, Q. He, Strength properties of geopolymers derived from original and desulfurized red mud cured at ambient temperature, Constr. Build. Mater. 125 (2016) 905–911.

[95] M. Zhang, M. Zhao, G. Zhang, D. Mann, K. Lumsden, M. Tao, Durability of red mud-fly ash based geopolymer and leaching behavior of heavy metals in sulfuric acid solutions and deionized water, Constr. Build. Mater. 124 (2016) 373–382.

[96] X. Ke, S.A. Bernal, N. Ye, J.L. Provis, J. Yang, One-part geopolymers based on thermally treated red Mud/NaOH blends, J. Am. Ceram. Soc. 98 (2015) 5–11.

[97] N. Ye, J. Yang, S. Liang, Y. Hu, J. Hu, B. Xiao, Q. Huang, Synthesis and strength optimization of one-part geopolymer based on red mud, Constr. Build. Mater. 111 (2016) 317–325.

[98] H. Choo, S. Lim, W. Lee, C. Lee, Compressive strength of one-part alkali activated fly ash using red mud as alkali supplier, Constr. Build. Mater. 125 (2016) 21–28.

[99] S. Chandrasekhar, K.G. Satyanarayana, P.N. Pramada, P. Raghavan, T.N. Gupta, Processing, properties and applications of reactive silica from rice husk - an overview, J. Mater. Sci. 38 (2003) 3159–3168.

[100] V.P. Della, I. Kühn, D. Hotza, Rice husk ash as an alternate source for active silica production, Mater. Lett. 57 (2002) 818–821.

[101] L. Armesto, A. Bahillo, K. Veijonen, A. Cabanillas, J. Otero, Combustion behaviour of rice husk in a bubbling fluidised bed, Biomass Bioenergy 23 (2002) 171–179.

[102] A. Hajimohammadi, J.S.J. van Deventer, Solid reactant-based geopolymers from rice hull ash and sodium aluminate, Waste Biomass Valorization (2016).

[103] P. Sturm, G.J.G. Gluth, H.J.H. Brouwers, H.-. Kühne, Synthesizing one-part geopolymers from rice husk ash, Constr. Build. Mater. 124 (2016) 961–966.

[104] H.K. Venkatanarayanan, P.R. Rangaraju, Material characterization studies on low- and high-carbon rice husk ash and their performance in portland cement mixtures, Adv. Civ. Eng. Mat. 2 (2013) 266–287.

[105] A. Peys, H. Rahier, Y. Pontikes, Potassium-rich biomass ashes as activators in metakaolin-based inorganic polymers, Appl. Clay Sci. 119 (2016) 401–409.

[106] P. Sturm, G.J.G. Gluth, S. Simon, H.J.H. Brouwers, H.-. Kühne, The effect of heat treatment on the mechanical and structural properties of one-part geopolymer-zeolite composites, Thermochim. Acta 635 (2016) 41–58.

[107] P. Sturm, S. Greiser, G.J.G. Gluth, C. Jäger, H.J.H. Brouwers, Degree of reaction and phase content of silica-based one-part geopolymers investigated using chemical and NMR spectroscopic methods, J. Mater. Sci. 50 (2015) 6768–6778.

[108] G.J.G. Gluth, C. Lehmann, K. Rübner, H.-. Kühne, Geopolymerization of a silica residue from waste treatment of chlorosilane production, Mater. Struct. 46 (2013) 1291–1298.

[109] N.A. Pambudi, R. Itoi, R. Yamashiro, B.Y.C.S.S. Syah Alam, L. Tusara, S. Jalilinasrabady, J. Khasani, The behavior of silica in geothermal brine from Dieng geothermal power plant, Indonesia, Geothermics 54 (2015) 109–114.

[110] A. Hajimohammadi, J.L. Provis, J.S.J. van Deventer, One-part geopolymer mixes from geothermal silica and sodium aluminate, Ind. Eng. Chem. Res. 47 (2008) 9396–9405.

[111] M.I. Khan, R. Siddique, Utilization of silica fume in concrete: review of durability properties, Resour. Conserv. Recycl. 57 (2011) 30–35.

[112] C. Li, H. Sun, L. Li, A review: The comparison between alkali-activated slag (Si Ca) and metakaolin (Si Al) cements, Cem. Concr. Res. 40 (2010) 1341–1349.

[113] M.S. Kim, Y. Jun, C. Lee, J.E. Oh, Use of CaO as an activator for producing a price-competitive non-cement structural binder using ground granulated blast furnace slag, Cem. Concr. Res. 54 (2013) 208–214.

[114] G. Lagaly, W. Tufar, A. Minihan, A. Lovell, Silicates, Ullmann's Encyclopedia of Industrial Chemistry, Wiley-VCH, Weinheim, 2000.

[115] M.X. Peng, Z.H. Wang, S.H. Shen, Q.G. Xiao, Synthesis, characterization and mechanisms of one-part geopolymeric cement by calcining low-quality kaolin with alkali, Mater. Struct. 48 (2015) 699–708.

[116] M.X. Peng, Z.H. Wang, S.H. Shen, Q.G. Xiao, L.J. Li, Y.C. Tang, L.L. Hu, Alkali fusion of bentonite to synthesize one-part geopolymeric cements cured at elevated temperature by comparison with two-part ones, Constr. Build. Mater. 130 (2017) 103–112.

[117] M. Kovtun, E.P. Kearsley, J. Shekhovtsova, Dry powder alkali-activated slag cements, Adv. Cem. Res. 27 (2015) 447–456.

[118] O. Helmboldt, L. Keith, H.C. Misra, K. Wefers, W. Heck, H. Stark, M. Danner, N. Rösch, Inorganic aluminum compounds, Ullmann's Encyclopedia of Industrial Chemistry, Wiley-VCH Verlag GmbH & Co. KGaA, 2000.

[119] D. Hardjito, S.E. Wallah, D.M.J. Sumajouw, B.V. Rangan, Fly ash-based geopolymer concrete, Aust. J. Struct. Eng. 6 (2005) 77–86.

[120] M. Palacios, P.F.G. Banfill, F. Puertas, Rheology and setting of alkali-activated slag pastes and mortars: effect if organic admixture, ACI Mater. J. 105 (2008) 140–148.

[121] M. Palacios, F. Puertas, Effect of superplasticizer and shrinkage-reducing admixtures on alkali-activated slag pastes and mortars, Cem. Concr. Res. 35 (2005) 1358–1367.

[122] D. Hardjito, S.E. Wallah, D.M.J. Sumajouw, B.V. Rangan, On the development of fly ash-based geopolymer concrete, ACI Mater. J. 101 (2004) 467–472.

[123] M. Palacios, Y.F. Houst, P. Bowen, F. Puertas, Adsorption of superplasticizer admixtures on alkali-activated slag pastes, Cem. Concr. Res. 39 (2009) 670–677.

[124] B. Nematollahi, J. Sanjayan, Effect of superplasticizers on workability of fly ash based geopolymer, in: R. Hassan, M. Yusoff, Z. Ismail, N. Amin, M. Fadzil (Eds.), InCIEC 2013, Springer, Singapore, 2014, pp. 713–719.

[125] F.A. Memon, M.F.L. Nuruddin, S. Demie, N. Shafiq, Effect of superplasticizer and extra water on workability and compressive strength of self-compacting geopolymer concrete, Res. J. Appl. Sci. Eng. Technol. 4 (2012) 407–414.

[126] L. Carabba, S. Manzi, M.C. Bignozzi, Superplasticizer addition to carbon fly ash geopolymers activated at room temperature, Mater. 9 (2016) 586.

[127] K.H. Yang, J.K. Song, J.S. Lee, Properties of alkali-activated mortar and concrete using lightweight aggregates, Mater. Struct. 43 (2010) 403–416.

[128] T. Suwan, M. Fan, Effect of manufacturing process on the mechanisms and mechanical properties of fly ash-based geopolymer in ambient curing temperature, Mater. Manuf. Process. 32 (2017) 461–467.

[129] N. Garg, C.E. White, Mechanism of zinc oxide retardation in alkali-activated materials: an in situ X-ray pair distribution function investigation, J. Mat. Chem. A. 5 (2017) 11794–11804.

[130] C. Gong, N. Yang, Effect of phosphate on the hydration of alkali-activated red mud–slag cementitious material, Cem. Concr. Res. 30 (2000) 1013–1016.

[131] J.J. Chang, A study on the setting characteristics of sodium silicate-activated slag pastes, Cem. Concr. Res. 33 (2003) 1005–1011.

[132] L. Kalina, V. Bílek, R. Novotnỳ, M. Mončeková, J. Másilko, J. Koplík, Effect of Na₃PO₄ on the hydration process of alkali-activated blast furnace slag, Mater. 9 (2016).

[133] A. Kusbiantoro, M.S. Ibrahim, K. Muthusamy, A. Alias, Development of sucrose and citric acid as the natural based admixture for fly ash based geopolymer, proc, Environ. Sci. 17 (2013) 596–602.

[134] A.R. Brough, M. Holloway, J. Sykes, A. Atkinson, Sodium silicate-based alkali-activated slag mortars. Part II. The retarding effect of additions of sodium chloride or malic acid, Cem. Concr. Res. 30 (2000) 1375–1379.

[135] A. Hajimohammadi, J.L. Provis, J.S.J. van Deventer, Time-resolved and spatially-resolved infrared spectroscopic observation of seeded nucleation controlling geopolymer gel formation, J. Colloid Interface Sci. 357 (2011) 384–392.

[136] B. Nematollahi, J. Sanjayan, Ambient temperature cured one-part engineered geopolymer composite: a sustainable alternative to engineered cementitious composite, 9th Rilem International Symposium on Fiber Reinforced Concrete, 2016 http://www.researchgate.net/publication/308645467_ambient_temperature_cured_one-part_engineered_geopolymer_composite_a_sustainable_alternative_to_engineered_cementitious_composites , Accessed date: 18 July 2017.

[137] B. Nematollahi, J. Sanjayan, Influence of type of fiber on tensile performance of one-part "dry-mix" strain hardening geopolymer composite (SHGC), in: K. Maekawa, A. Kasuga, J. Yamazaki (Eds.), Proceedings of 11th Fib International PhD Symposium in Civil Engineering, Technical University of Tokyo, Tokyo, 2016, pp. 831–838.

[138] B. Nematollahi, J. Sanjayan, F.U. Ahmed S, Tensile strain hardening behavior of PVA fiber-reinforced engineered geopolymer composite, J. Mater. Civ. Eng. 27 (2015) 04015001.

[139] B. Nematollahi, J. Sanjayan, F.U.A. Shaikh, Comparative deflection hardening behavior of short fiber reinforced geopolymer composites, Constr. Build. Mater. 70 (2014) 54–64.

[140] I. García-Lodeiro, O. Maltseva, A. Palomo, A. Fernández-Jiménez, Hybrid alkaline cements. Part I: Fundamentals, Rev. Rom. Mater. Rom. J. Mater. 42 (2012) 330–335.

[141] C. Shi, A.F. Jiménez, A. Palomo, New cements for the 21st century: the pursuit of an alternative to Portland cement, Cem. Concr. Res. 41 (2011) 750–763.

[142] I. García-Lodeiro, A. Fernández-Jiménez, A. Palomo, Variation in hybrid cements over time. Alkaline activation of fly ash-portland cement blends, Cem. Concr. Res. 52 (2013) 112–122.

[143] I. Garcia-Lodeiro, S. Donatello, A. Fernández-Jiménez, Á. Palomo, Hydration of hybrid alkaline cement containing a very large proportion of fly ash: a descriptive model, Mater. 9 (2016).

[144] A. Palomo, A. Fernández-Jiménez, G. Kovalchuk, L.M. Ordoñez, M.C. Naranjo, Opc-fly ash cementitious systems: Study of gel binders produced during alkaline hydration, J. Mater. Sci. 42 (2007) 2958–2966.

[145] I. Garcia-Lodeiro, V. Carcelen-Taboada, A. Fernández-Jiménez, A. Palomo, Manufacture of hybrid cements with fly ash and bottom ash from a municipal solid waste incinerator, Constr. Build. Mater. 105 (2016) 218–226.

[146] A. Fernández-Jiménez, I. García-Lodeiro, S. Donatello, O. Maltseva, A. Palomo, Specific examples of hybrid alkaline cement, MATEC Web Conf. 11 (2014).

[147] I. García-Lodeiro, A. Fernández-Jimenez, A. Palomo, Cements with a low clinker

content: Versatile use of raw materials, J. Sustain. Cement-Based Mater. 4 (2015) 140–151.

[148] Z. Abdollahnejad, P. Hlavacek, S. Miraldo, F. Pacheco-Torgal, J.L.B. De Aguiar, Compressive strength, microstructure and hydration products of hybrid alkaline cements, Mater. Res. 17 (2014) 829–837.

[149] Z. Abdollahnejad, Development of Foam One-Part Geopolymers (Ph.D. Thesis), Minho University, Portugal, 2016.

[150] Z. Abdollahnejad, C.M.G. Jesus, F.P. Torgal, J.L. Aguiar, One-part geopolymers versus Ordinary Portland Cement (OPC) mortars: durability assessment, http://hdl.handle.net/1822/26202, (2013) , Accessed date: 18 July 2017.

[151] S. Sperinck, P. Raiteri, N. Marks, K. Wright, Dehydroxylation of kaolinite to metakaolin—a molecular dynamics study, J. Mater. Chem. 21 (2011) 2118–2125.

[152] M.X. Peng, Z.H. Wang, Q.G. Xiao, F. Song, W. Xie, L.C. Yu, H.W. Huang, S.J. Yi, Effects of alkali on one-part alkali-activated cement synthesized by calcining bentonite with dolomite and Na₂CO₃, Appl. Clay Sci. 139 (2017) 64–71.

[153] D. Koloušek, J. Brus, M. Urbanova, J. Andertova, V. Hulinsky, J. Vorel, Preparation, structure and hydrothermal stability of alternative (sodium silicate-free) geopolymers, J. Mater. Sci. 42 (2007) 9267–9275.

[154] S.J. O'Connor, K.J.D. MacKenzie, Synthesis, characterisation and thermal behaviour of lithium aluminosilicate inorganic polymers, J. Mater. Sci. 45 (2010) 3707–3713.

[155] S. Lee, A. van Riessen, C.-. Chon, Benefits of sealed-curing on compressive strength of fly ash-based geopolymers, Mater. 9 (2016).

[156] P. Duxson, S.W. Mallicoat, G.C. Lukey, W.M. Kriven, J.S.J. van Deventer, The effect of alkali and Si/Al ratio on the development of mechanical properties of metakaolin-based geopolymers, Colloids Surf. A Physicochem. Eng. Asp. 292 (2007) 8–20.

[157] P. Duxson, J.L. Provis, G.C. Lukey, S.W. Mallicoat, W.M. Kriven, J.S.J. Van Deventer, Understanding the relationship between geopolymer composition, microstructure and mechanical properties, Colloids Surf. A Physicochem. Eng. Asp. 269 (2005) 47–58.

[158] A. Hajimohammadi, T. Ngo, P. Mendis, T. Nguyen, A. Kashani, J.S.J. van Deventer, Pore characteristics in one-part mix geopolymers foamed by H₂O₂: the impact of mix design, Mater. Des. 130 (2017) 381–391.

[159] G.J.G. Gluth, C. Lehmann, K. Rübner, H.C. Kühne, Geopolymerization of a silica residue from waste treatment of chlorosilane production, Mater. Struct. 46 (2013) 1291–1298.

[160] P. Hewlett, Lea's Chemistry of Cement and Concrete, Butterworth-Heinemann, Oxford, 2003.

[161] A. Hajimohammadi, J.L. Provis, J.S.J. van Deventer, Effect of alumina release rate on the mechanism of geopolymer gel formation, Chem. Mater. 22 (2010) 5199–5208.

[162] K.H. Yang, J.K. Song, K.S. Lee, A.F. Ashour, Flow and compressive strength of alkali-activated mortars, ACI Mater. J. 106 (2009) 50–58.

[163] H.A. Abdel-Gawwad, S.A. Abo-El-Enein, A novel method to produce dry geopolymer cement powder, HBRC J. 12 (2016) 13–24.

[164] EN 206-1, Concrete–Part 1: Specification, Performance, Production and Conformity, CEN, Brussels, 2000.

[165] ASTM, C1437–15, Standard Test Method for Flow of Hydraulic Cement Mortar, ASTM International, West Conshohocken, PA, 2015.

[166] EN 197-1, Cement - Part 1: Composition, Specifications and Conformity Criteria for Common Cements, (2000).

[167] C. Rees, Mechanisms and Kinetics of Gel Formation in Geopolymers (Ph.D. Thesis), University of Melbourne, 2007.

[168] C.A. Rees, J.L. Provis, G.C. Lukey, J.S.J. van Deventer, The mechanism of geopolymer gel formation investigated through seeded nucleation, Colloids Surf. A Physicochem. Eng. Asp. 318 (2008) 97–105.

[169] S.A. Bernal, J.L. Provis, A. Fernández-Jiménez, P.V. Krivenko, E. Kavalerova, M. Palacios, C. Shi, Binder chemistry - High-calcium alkali-activated materials, in: J.L. Provis, J.S.J. Van Deventer (Eds.), Alkali Activated Materials, State-of-the-Art Report, RILEM TC 224-AAM, Springer, Dordrecht, 2014, pp. 59–91.

[170] J.L. Provis, A. Fernández-Jiménez, E. Kamseu, C. Leonelli, A. Palomo, Binder chemistry – Low-calcium alkali-activated materials, Anonymous Alkali Activated Materials, State-of-the-Art Report, RILEM TC 224-AAM, Springer, Dordrecht, 2014, pp. 93–123.

[171] A. Hajimohammadi, J.L. Provis, J.S.J. van Deventer, The effect of silica availability on the mechanism of geopolymerisation, Cem. Concr. Res. 41 (2011) 210–216.

[172] R.J. Myers, S.A. Bernal, J.L. Provis, A thermodynamic model for C-(N-)A-S-H gel: CNASH_ss. Derivation and validation, Cem. Concr. Res. 66 (2014) 27–47.

[173] Q. Wan, F. Rao, S. Song, R.E. García, R.M. Estrella, C.L. Patiño, Y. Zhang, Geopolymerization reaction, microstructure and simulation of metakaolin-based geopolymers at extended Si/Al ratios, Cem. Concr. Compos. 79 (2017) 45–52.

[174] R.J. Myers, S.A. Bernal, R. San Nicolas, J.L. Provis, Generalized structural description of calcium-sodium aluminosilicate hydrate gels: The cross-linked substituted tobermorite model, Langmuir 29 (2013) 5294–5306.

[175] B. Walkley, R.S. Nicolas, S. Marc-Antoine, G.J. Rees, J.V. Hanna, J.S.J. van Deventer, J.L. Provis, Phase evolution of C-(N)-A-S-H/N-A-S-H gel blends investigated via alkali-activation of synthetic calcium aluminosilicate precursors, Cem. Concr. Res. 89 (2016) 120–135.

[176] I. García-Lodeiro, A. Palomo, A. Fernández-Jiménez, D.E. Macphee, Compatibility studies between N-A-S-H and C-A-S-H gels. Study in the ternary diagram Na₂O–CaO–Al₂O₃–SiO₂–H₂O, Cem. Concr. Res. 41 (2011) 923–931.

[177] I. García-Lodeiro, A. Fernandez-Jimenez, A. Palomo, Hydration kinetics in hybrid binders: Early reaction stages, Cem. Concr. Compos. 39 (2013) 82–92.

[178] I.S.O. 14040, Environmental Management, Life Cycle Assessment, Principles and Framework, International Organization for Standardization, Geneva, 2006.

[179] J. Rockström, W. Steffen, K. Noone, Å. Persson, F.S. Chapin, E.F. Lambin, T.M. Lenton, M. Scheffer, C. Folke, H.J. Schellnhuber, B. Nykvist, C.A. De Wit, T. Hughes, S. Van Der Leeuw, H. Rodhe, S. Sörlin, P.K. Snyder, R. Costanza, U. Svedin, M. Falkenmark, L. Karlberg, R.W. Corell, V.J. Fabry, J. Hansen, B. Walker, D. Liverman, K. Richardson, P. Crutzen, J.A. Foley, A safe operating space for humanity, Nature 461 (2009) 472–475.

[180] G. Habert, J.B. D'Espinose De, N. Lacaillerie, Roussel, An environmental evaluation of geopolymer based concrete production: Reviewing current research trends, J. Clean. Prod. 19 (2011) 1229–1238.

[181] L.K. Turner, F.G. Collins, Carbon dioxide equivalent (CO2-e) emissions: A comparison between geopolymer and OPC cement concrete, Constr. Build. Mater. 43 (2013) 125–130.

[182] E. Jamieson, B. McLellan, A. Van Riessen, H. Nikraz, Comparison of embodied energies of Ordinary Portland Cement with Bayer-derived geopolymer products, J. Clean. Prod. 99 (2015) 112–118.

[183] B.C. McLellan, R.P. Williams, J. Lay, A. Van Riessen, G.D. Corder, Costs and carbon emissions for geopolymer pastes in comparison to ordinary portland cement, J. Clean. Prod. 19 (2011) 1080–1090.

[184] A. Mellado, C. Catalán, N. Bouzón, M.V. Borrachero, J.M. Monzó, J. Payá, Carbon footprint of geopolymeric mortar: study of the contribution of the alkaline activating solution and assessment of an alternative route, RSC Adv. 4 (2014) 23846–23852.

[185] A. Petrillo, R. Cioffi, F. De Felice, F. Colangelo, C. Borrelli, An environmental evaluation: a comparison between geopolymer and OPC concrete paving blocks manufacturing process in italy, Environ. Prog. Sustain. Energy 35 (2016) 1699–1708.

[186] A. Heath, K. Paine, M. McManus, Minimising the global warming potential of clay based geopolymers, J. Clean. Prod. 78 (2014) 75–83.

[187] M. Weil, K. Dombrowski, A. Buchwald, Life-cycle analysis of geopolymers, in: J.L. Provis, J.S.J. van Deventer (Eds.), Geopolymers: Structures, Processing, Properties and Industrial Applications, CRC Press, Boca Raton, 2009, pp. 194–210.

[188] Z. Abdollahnejad, S. Miraldo, F. Pacheco-Torgal, J.B. Aguiar, Cost-efficient one-part alkali-activated mortars with low global warming potential for floor heating systems applications, Eur. J. Environ. Civ. Eng. 21 (2017) 412–429.

[189] K.-. Yang, J.-. Song, K.-. Song, Assessment of CO 2 reduction of alkali-activated concrete, J. Clean. Prod. 39 (2013) 265–272.

[190] K.-. Yang, K.-. Lee, J.-. Song, M.-. Gong, Properties and sustainability of alkali-activated slag foamed concrete, J. Clean. Prod. 68 (2014) 226–233.

[191] G. Habert, C. Ouellet-Plamondon, Recent update on the environmental impact of geopolymers, RILEM Tech. Lett. 1 (2016) 17–23.

[192] C. Ouellet-Plamondon, G. Habert, Life cycle assessment (LCA) of alkali-activated cements and concretes, in: F. Pacheco-Torgal, J. Labrincha, C. Leonelli, A. Palomo, P. Chindaprasit (Eds.), Handbook of Alkali-Activated Cements, Mortars and Concretes, Elsevier, Amsterdam, 2014, pp. 663–686.

[193] J. Davidovits, Environmental implications of Geopolymers, https://www.materialstoday.com/polymers-soft-materials/features/environmental-implications-of-geopolymers/, (2017) , Accessed date: 17 July 2017.

[194] M. Fawer, M. Concannon, W. Rieber, Life cycle inventories for the production of sodium silicates, Int. J. Life Cycle Assess. 4 (1999) 207–212.

[195] C.C.S. Chan, D. Thorpe, M. Islam, An evaluation of life long fly ash based geopolymer cement and ordinary Portland cement costs using extended life cycle cost method in Australia, 2015 IEEE International Conference on Industrial Engineering and Engineering Management (IEEM), IEEE, Singapore, 2016, , http://dx.doi.org/10.1109/IEEM.2015.7385607.