

RINOPOLYCRETE - TOWARDS A CEMENT-FREE AND FULLY RECYCLED CONCRETE



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(RInoPolyCrete)

Task 1 - Report 2

Database on alkali-activated materials containing municipal solid waste incinerator bottom ash

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Table of contents

1. Introduction	1
2. Methodology	2
2.1. Literature search	2
2.2. Search results and appraisal	4
2.3. Selected criteria	6
2.3.1. Oxides composition of precursor	7
2.3.2. Alkali activator	8
2.3.3. Compositions	10
2.3.4. Curing conditions	10
3. Performance of MIBA-based alkali-activated materials	10
4. Final remarks	15
References	16

List of Figures

Figure 1 - Number of studies per (a) country of study, (b) year of study, (c) material type, (d) publisher, (e) temperature of curing, (f) type of alkali activator, (g) type of precursor and (h) studied parameter	5
Figure 2 - Oxides composition of MIBA from various studies.....	11
Figure 3 - Compressive strength vs. (a) MIBA incorporation ratio, (b) liquid/solid ratio, (c) total SiO ₂ quantity in solution, (d) NaOH concentration and (e) mix duration	13
Figure 4 - Effect of various types of precursor on AAM	14
Figure 5 - Density of AAM vs. (a) MIBA incorporation ratio and (b) liquid to solid ratio	14
Figure 6 - Relationship between density and (a) compressive strength and (b) porosity of AAM.....	15

List of Tables

Table 1 - Search options and databases	3
Table 2 - Main criteria considered for each selected study	6
Table 3 - Chemical composition of all types of Portland cement	7
Table 4 - Influence of chemical activator on the performance of alkali-activated concrete	9
Table 5 - Calculating the total amount of Na ₂ O, SiO ₂ and H ₂ O of alkali activators.....	10

Acronyms:

AAM - alkali activated materials

FA - fly ash;

GGBS - ground granulated blast furnace slag;

MIBA - municipal solid waste incinerator bottom ash;

MIFA - municipal solid waste incinerated fly ash;

OPC - ordinary Portland cement;

SCM - supplementary cementitious materials.

1. Introduction

This report presents the database on alkali-activated materials containing municipal solid waste incinerator bottom ash that has been prepared for Task 1 within the scope of the FCT Project ECI-COM/29196/2017- “Recycled inorganic polymer concrete - Towards a cement-free and fully recycled concrete (RInoPolyCrete)”. The main aim of this database is to analyse the literature on alkali-activated materials containing municipal solid waste incinerator bottom ash and draw conclusions that can be helpful in the subsequent tasks of the project.

One way to promote sustainability is by utilizing co-products or by-products as partial replacement of cement. However, their incorporation ratios are limited because, after a given ratio (high volume), further hydration products in the paste may not be produced. To overcome this issue, alkaline activator can be used. Thus, alkali activation technique can be considered an alternative process for partial replacement of supplementary cementitious materials (SCM). There are several materials that can be used as precursors, e.g. municipal solid waste incinerator bottom ash (MIBA), fly ash (FA), municipal incinerated fly ash (MIFA) and ground granulated blast furnace slag (GGBS). In other words, materials that are rich in amorphous Al_2O_3 and SiO_2 can be used as a precursor. The mentioned oxide materials dissolve and undergo polymerization with the use of strong alkaline solutions (e.g. NaOH, KOH and Na_2SiO_3), and form a “three-dimensional amorphous aluminosilicate” network. In this database, we mainly focused on MIBA due to the reasons described in a study of Kurda et al. [1], namely promoting sustainability.

One way to understand the effect of alkali-activated MIBA is by preparing a systematic and extensive database to synthesize, identify and evaluate the existing research on the mentioned topic. Therefore, this work is mainly focused on how the database (Appendix I) was made, namely the selected studies and parameters. Also, the criteria to handle missing data are explained. In addition, a preliminary analysis is made of the main parameters in order to understand the general trends on the mentioned topic and identify future developments.

2. Methodology

2.1. Literature search

This work is a systematic and extensive plan to synthesize, identify, and evaluate the literature from the state-of-the-art report made by the same authors [1]. Thereafter, this work is followed by an exhaustive analysis of the literature to identify topics for further study. This work is mainly focused on the properties (e.g. mechanical and durability-related) of alkali-activated materials (paste, mortar and concrete) containing MIBA.

In order to analyse the performance of alkali-activated materials (AAM) containing MIBA, a literature research was made using the search engines of several databases and “Google” (Table 1). For each database, the same search options were repeated using combinations of the following keywords:

- Bottom ash;
- Geopolymer precursor;
- Geopolymer;
- Hybrid cements;
- Hydrothermal synthesis;
- IBA (Incinerator bottom ash);
- Incineration;
- Incinerator bottom ash;
- Inertization and valorisation of ashes by alkali activation;
- MIBA (Municipal solid waste incinerator bottom ash);
- MSW (Municipal solid waste);
- MSWI (Municipal solid waste incinerator);
- Municipal incinerator bottom ash;
- Municipal solid waste incineration ash slag;

- Stabilization/solidification of a municipal solid waste incineration;
- Synthesis;
- Waste management;
- Waste plant.

Table 1 - Search options and databases

Main database *	Search options (for all databases)
Google Scholar	Data range: from past to May, 2019
ScienceDirect	Search in: Any, topic, title, articles with author supplied keywords
Research Gate	Document type: Article, Data Paper, Review
Taylor and Francis	Language: Any
Web of Science	Article types. Review articles, research articles, data paper
Scopus	

* Beside of the main database, other database are also considered (e.g. ICE, Wiley Online Library, RILEM, Web of Knowledge)

Serval boundaries were made to find the related studies for further inspection. The validity of the selected papers was specified by analysing the title, abstract, materials and methodology of the research studies. Thereafter, the non-relevant studies were removed (e.g. studies related to AAM containing MIBA as an aggregate). For that purpose, two main criteria were prepared in order to demonstrate whether a material is relevant to this research work. The chosen studies must meet the following criteria:

- The objective of this review report is to find the papers pertaining to AAM made as paste, mortar and concrete made with MIBA only or MIBA blended with other SCM or ordinary Portland cement (OPC). In addition, the studies without alkali-activation solution are excluded;
- MIBA must be used as a precursor (binder). In other words, the other applications of MIBA i.e. studies on AAM containing MIBA as an aggregate are excluded. The reason behind the selected application is that binder (e.g. cement) can be considered the main contributor to the environmental impact of the construction materials.

In addition, the main focus of this study is on the use of MIBA without any thermal treatments. This is related to the environmental impact because thermal treatment consumes a significant

amount of energy. Thus, the technique may not be considered as an environmentally friendly solution. However, studies that used thermal treatment are not excluded due to the limited number of studies using untreated MIBA. Thus, the results of both options are compared.

2.2. Search results and appraisal

According to the search process made in the previous sub-section (§2.1), the basic body of the literature comprised 22 studies [2-23]. Most of the studies were carried out in Asia and the others were made in Europe (Figure 1a). In addition, the “publication year” of the mentioned studies ranges from 2003-2019 and most of the studies were made in 2016-2019 (Figure 1b). Furthermore, most of the studies were related to the performance of alkali-activated paste [2-15] and mortar [16-22], and only one study [23] was related to concrete (Figure 1c). All the references are classified as a “case study”, and the majority of them are from journals, namely Construction and Building Materials, Journal of Cleaner Production, Cement and Concrete Composite and Waste Management (Figure 1d).

Regarding the experimental campaign, most researchers cured the samples at 60-80 °C for AAM made only with MIBA. Others used room temperature for the AAM made with MIBA and other SCM or OPC (Figure 1e). Concerning the alkali-activators, the majority of the studies utilized only NaOH or Na₂SiO₃ mixed with NaOH (Figure 1f). In addition, the type of precursors is mainly MIBA. However, some studies also incorporated OPC and several types of SCM with MIBA to improve the performance of AAM (Figure 1g). Generally, the results are related to the compressive strength (17 studies) and toxicity (11 studies) of AAM containing MIBA. Regarding the durability performance, only the microstructure characteristics are considered as a main parameter (Figure 1h).

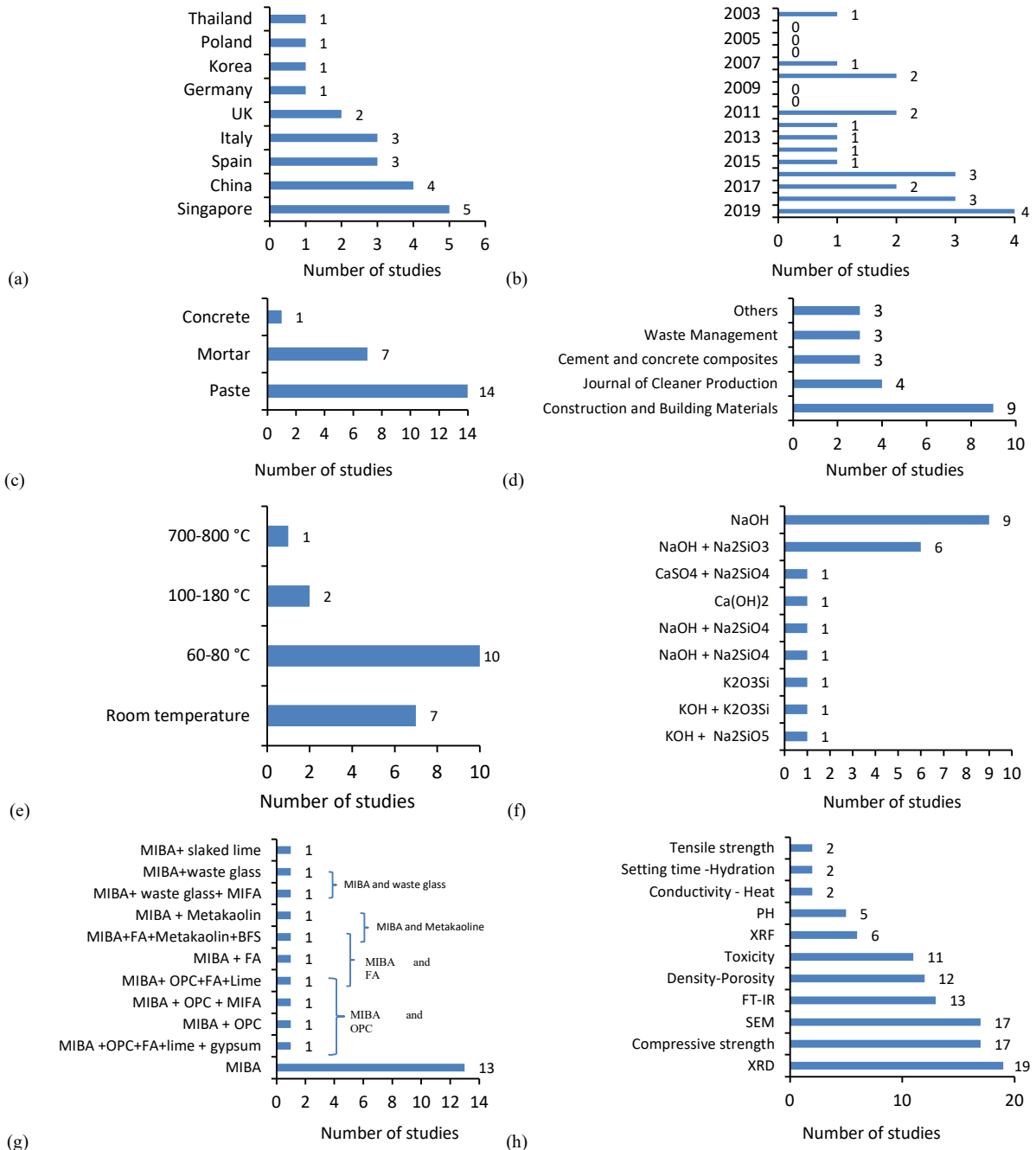


Figure 1 - Number of studies per (a) country of study, (b) year of study, (c) material type, (d) publisher, (e) temperature of curing, (f) type of alkali activator, (g) type of precursor and (h) studied parameter

2.3. Selected criteria

In this study, the following main criteria (Table 2) were considered for each study found in section 2.2 in order to understand the effect of MIBA on AAM. Apart from the mentioned criteria, several other secondary criteria were considered, namely for the paste, mortar and concrete properties. However, they are not mentioned in the table because the selected studies (Figure 2) did not investigate them (e.g. carbonation, chloride ion penetration and water absorption). Each main criterion is explained in depth in the following sub-sections (§2.3.1-2.3.5).

Table 2 - Main criteria considered for each selected study

Authors name	Description of publication
Oxides composition of precursor (% by weight)	SiO ₂ ; Al ₂ O ₃ ; CaO; Fe ₂ O ₃ ; K ₂ O; MgO; SO ₃ ; TiO ₂ ; P ₂ O ₅ ; Na ₂ O; ZnO; CuO; PbO; Cl; Loss on ignition (LOI).
Alkali activator	Solution (S) type S1 concentration of solution (M) S2 concentration (M) Weight of solution Mass ratio of the solutions SiO ₂ (%weight of total solution) Na ₂ O _{eq} (%weight of total solution) CaO _{eq} (%weight of total solution) Water (%weight of total solution) SiO ₂ /Na ₂ O _{eq} Density 25 °C, g/cm ³ Si/Al Na/Al Other solution
Composition	Percentage of precursor (% by weight) Precursor content (kg/m ³) Mass ratio of precursors Total precursor content - Solid (kg/m ³) Total percentage (% by weight) Solution - Liquid (kg/m ³) Total water (% by weight) Total water content (kg/m ³) L/S ratio Fine natural aggregates < 4 mm (kg/m ³) Water reducing admixture type Water reducing admixture (kg/m ³)
Curing conditions	Temperature (°C) Relative humidity (%) Period (hours) Additional curing
Paste, Mortar and concrete properties	Workability Compressive strength Flexural and tensile strength (MPa) Modulus of elasticity (GPa) Dry density (kg/m ³) Porosity (%)

2.3.1. Oxides composition of precursor

It is known that the oxide composition, namely SiO_2 , Al_2O_3 , CaO , Fe_2O_3 , K_2O , MgO , SO_3 , TiO_2 , P_2O_5 , Na_2O , ZnO , CuO , PbO , and Cl , and Loss on ignition (LOI) play a vital role in identifying the suitability of any precursor. In other words, they give an idea about the precursor that can be used as binder or filler.

As a benchmark for other types of binder/precursor, Kosmatka et al. [24] determined the oxide composition for all the types of Portland cement, and identified minimum, maximum and mean values for the main chemical composition (Table 3). It is important to know the quantity of CaO and SiO_2 in any precursor because they react and produce C-S-H (major contributor to the strength of AAM). Al_2O_3 may not contribute to the strength of the paste. However, it is the one of the main contributor to form monosulphate [25], which increases the durability performance of the AAM [26, 27].

Table 3 - Chemical composition of all types of Portland cement

All types of Portland cement	Chemical composition (%)						
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	$\text{Na}_2\text{O eq}$
Min-Max	18.6-24.4	2.2-7.3	0.2-5.9	61.3-68.7	0.3-4.5	1.7-4.9	0.09-1.2
Mean	21.3	4.5	3.0	63.9	2.0	2.8	0.5

Na_2O and K_2O can be referred to as alkalis activators of MIBA. They react with active SiO_2 of the aggregates and form alkali-silica gel. The gel can expand and cause cracking and internal failure at longer ages in conventional concrete. Generally, the percentage of the abovementioned alkalis must not be higher than 1% for OPC concrete [28]. However, such limit is not imposed for AAM, since their presence is vital for the alkali reactions to occur.

It may not be reliable to compare the threshold values defined for the chemical composition of cement and those of any type of precursor used to produce AAM. However, the threshold values of cement can be used as a benchmark and provide a preliminary idea about the performance of the precursor. For example, according to EN-197-1 [29], the percentage of SO_3 in

cement must be up to 4.5% as high contents lead to internal disruption. However, this limit may not make sense in AAM, since these are much more resilient to sulphate attack. The range of MgO in cement should be around 1-4% in order to control the expansion during the hydration process, but, in AAM, the mechanism of MgO hydration and consequent expansion is not entirely known and thus the limit for this constituent may be further increased. Furthermore, TiO₂ is a photo-catalytic component and may not directly affect the chemical reaction of the paste [30]. Moreover, P₂O₅ in cement decreases the early strength of the paste when its content is higher than 2.25% of clinker mass because it reacts with 2CaO and SiO₂ and decreases C-S-H as a result. Normally, CuO and ZnO need to be determined in order to understand the risk of leaching potential of paste due to Cu and Zn metals. Additionally, ZnO, CuO and PbO can be considered as retarders (delaying the hydration). Their mass concentration should be less than 0.3% [31], 1% [32] and 0.04% [33-35], respectively, for conventional concrete, but additional information is required for AAM.

2.3.2. Alkali activator

An alkali activator is used to react with the precursor (solid aluminosilicate) under alkaline conditions to produce hardened AAM, which is made in a complex “alkali-alkali earth-aluminosilicate and/or hydrous alkali-aluminosilicate” phase [36]. MOH and M₂SiO₃ (M is either K or Na) are the most commonly used activators. Other activators are also used, such as the Na₂CO₃ and Na₂SO₄ [37]. CaO, Ca(OH)₂ and MgO are also identified as potential activators, though less used [37].

There are two main procedures to produce AAM, either by dry-activator (dry activator - e.g. NaOH - mixed with water and precursor at the same time) or liquid-activator (the activator is already prepared and then mixed with precursor). In the database, both procedures are used in the selected studies. Provis [36] stated that the normal range of alkali component (activator without water) is 5-10% of total mass of the binder and the molar concentration of NaOH can be between 2 M and 14 M [38].

Generally, the influence of the alkali activator on AAM depends on its quantity and concentration and the precursor's reactivity (Table 4). Since the type of alkali activator significantly affects the performance of AAM, this parameter is considered one of the most vital. As shown in Table 4, the concentration of the activator and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio were considered in the database. It is known that a higher $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio decreases the workability because of the higher viscosity of Na_2SiO_3 . In addition, higher concentrations of NaOH increase the setting time of AAM [38].

Table 4 - Influence of chemical activator on the performance of alkali-activated concrete

Studies	Precursor *	$\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio, SS/SH	Optimal $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio	NaOH concentration (M)	Optimal NaOH (M)	Silica modulus	Optimal Silica modulus	Compressive strength (MPa)
Gorhan and Kurklu [39]	FA	0.4-2.3	-	3-9	6	-	-	12-23
Sukmak et al. [40]	FA	-	0.7	10	-	-	-	4-14
Somna et al. [41]	FA	0.33-3.0	-	4.5-16.5	-	-	-	7-25
Ridtirud et al. [42]	FA	7.5-12.5	1.5	-	7.5	-	-	25-45
Guo et al. [43]	FA	-	-	-	-	1.0-2.0	1.5	5-63
Law et al. [44]	FA	-	-	10	-	0.75-1.25	1.0	39-57
He et al. [45]	RHA	2.5	-	2-6	2	-	-	8-15
Nazari et al. [46]	RHA	-	-	4-12	12	-	-	20-30
Songpiriyakij et al. [47]	RHA	0.5-2.5	-	14, 18	18	-	-	34-56
Detphan and Chindapasirt [48]	RHA	1.9-5.5	4.0	-	-	-	-	15-40
Salih et al. [49]	POFA	0.5-3.0	2.5	10	-	-	-	7-32
Yusuf et al. [50]	POFA	-	-	10	-	0.92-1.64	0.92	65-69
Ahmari and Zhang [51]	MS	-	-	10-15	15	-	-	4-34
Wongsa et al. [19]	MIBA	1	-	10	-	-	-	10.6
Zhu et al. [4]	MIBA	0.5	-	8	-	-	-	2.8

* FA - fly ash; RHA - rice husk ash; POFA - Palm oil fuel ash-based; MS - Hematite mine tailings

It is important to know the $\text{SiO}_2/\text{Na}_2\text{O}_{\text{eq}}$ ratio because both the relative amount and polymerization degree of the silicate gel phase are significantly affected by it (they decrease with decreasing $\text{SiO}_2/\text{Na}_2\text{O}_{\text{eq}}$ ratio [2]). For that purpose, SiO_2 and $\text{Na}_2\text{O}_{\text{eq}}$ (% by weight of total solution) were calculated by summing the quantity of each alkaline-activator (Table 5

Table 5).

Table 5 - Calculating the total amount of Na₂O, SiO₂ and H₂O of alkali activators

Components	NaOH			Na ₂ SiO ₃			Added water		
	Total weight (g)	%	Weight (g) ^a	Total weight (g)	%	Weight (g)	Total weight (g)	%	Weight (g)
Na ₂ O		N1 %	S1 *N1/(100*0.775) +		N2 %	S2*N3/100 +		0	0
SiO ₂	S1	0	0	S2	N3 %	S2*N4/100 +	S3	0	0
H ₂ O		100-N1 %	(100-N1) *N2/100 +		100-N2-N3 %	S2*(100-N2-N3)/100 +		100 %	M3*100/100

^a The amount of Na₂O from NaOH are defined to be of 77.5% of NaOH's mass. However, this value may slightly change due to extra water of the mixes at reverse reaction.

2.3.3. Compositions

It is known that the type and quantity of the precursors, water and aggregates, water reducing admixture and alkaline activator significantly affect the performance of AAM and thus are also considered in the database. Some studies showed the quantity of the mentioned materials by percentage and the others by mass. Therefore, the total percentage (% by weight) of precursor, namely for MIBA, are calculated (if not given). Then, liquid to solid ratios are determined.

2.3.4. Curing conditions

Curing conditions can be considered one of the most important parameters of any AAM. Normally, AAM samples are subjected to two types of curing. After demoulding, they are subjected to a relatively high curing temperature for a short period of time (generally, 60-80 °C for 10-72 hours), followed by curing at room temperature until the testing age. In this database, the focus is on the first step of the curing process, because it significantly affects the performance of AAM. The database also included the mixing duration and some additional techniques used by other researchers to improve the performance of AAM, e.g. pressing the samples at fresh state to remove the air bubbles and saturated steam pressure.

3. Performance of MIBA-based alkali-activated materials

In this section, the results presented in the Appendix I are simply explained in order to have an idea about the effect of the criteria mentioned in section 2 on the performance of AAM made

with MIBA. Apart from the fresh and hardened states of AAM containing MIBA, the physical (e.g. size of particles) and chemical characteristics (e.g. oxides composition) of MIBA are also very important criteria to understand the influence of MIBA on AAM. For example, the results show that there is a big scatter in the oxide compositions of MIBA samples. However, the sum of the three main oxides (SiO_2 , CaO and Al_2O_3) can be around 71% of the total oxide content of MIBA. Thus, MIBA may have potential to be used as an active precursor for AAM.

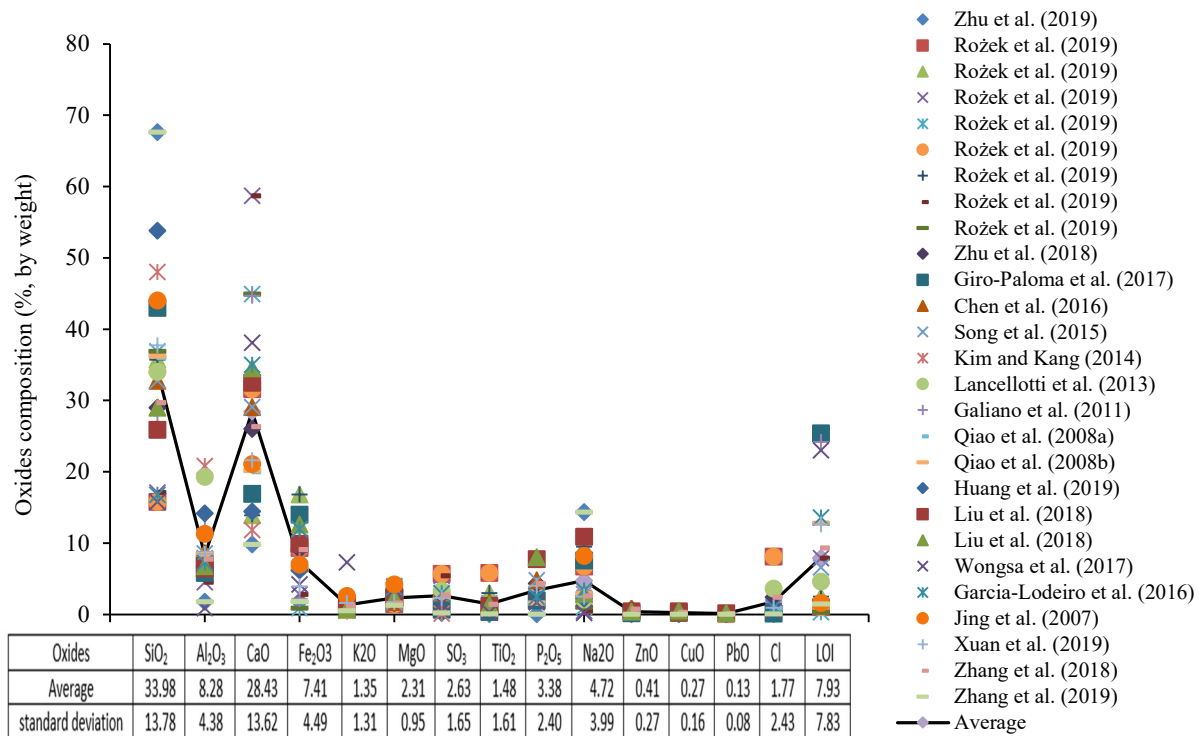


Figure 2 - Oxides composition of MIBA from various studies

As mentioned in section 2, the early age performance (e.g. slump and setting time) of AAM for all the selected studies are considered in the database. However, most of the studies did not focus on this state. Regarding the hardened state, even though there were some attempts to understand the compressive strength of alkali-activated MIBA, there are not many results. Figure 3a shows that the AAM can be produced with extremely varying compressive strength (0.3-75

MPa) regardless of the incorporation ratio of MIBA. According to a 95% confidence interval (excluding the AAM made with FA and metakaolin, and AAM mixes without modifying $\text{SiO}_2/\text{Na}_2\text{O}$), the strength of AAM seems to have decreased with increasing amount of MIBA.

The strength of AAM depends on several factors e.g. liquid to solid ratio (Figure 3b). The results show that the liquid to solid ratio of 0.5 can be considered as an optimum as the highest compressive strength were achieved for that value. Still, given the relatively low number of studies and their representativeness, further information should be gathered to ascertain this trend.

The quantity and characteristics of the alkali activator also play a major role on the performance of AAM (Figure 3c and d). The mixing duration is also one of the factors that affects the performance of AAM because it drives the materials to react and form polymer chains and release the air voids (i.e. hydrogen release). Figure 3e shows that the strength can be increased with increasing mix duration.

As mentioned, apart from the alkali activator, the type of the precursor is one of the main factors that affect the strength of AAM (Figure 4). The results show that the strength of alkali activated MIBA can be significantly increased by incorporating one of the following materials: OPC; MIFA; GBFS; and FA.

Knowledge regarding the durability of MIBA-based AAM is also very limited; most researchers have focused on the density and porosity. Generally, the density of alkali-activated MIBA is relatively low ($612\text{-}1036 \text{ kg/m}^3$) because of the air voids generated during the reaction of precursor (i.e. corrosion of metallic aluminium) and the alkaline solution (Figure 5a). As expected, the density of AAM mixes decreases with increasing liquid to solid ratio (Figure 5b).

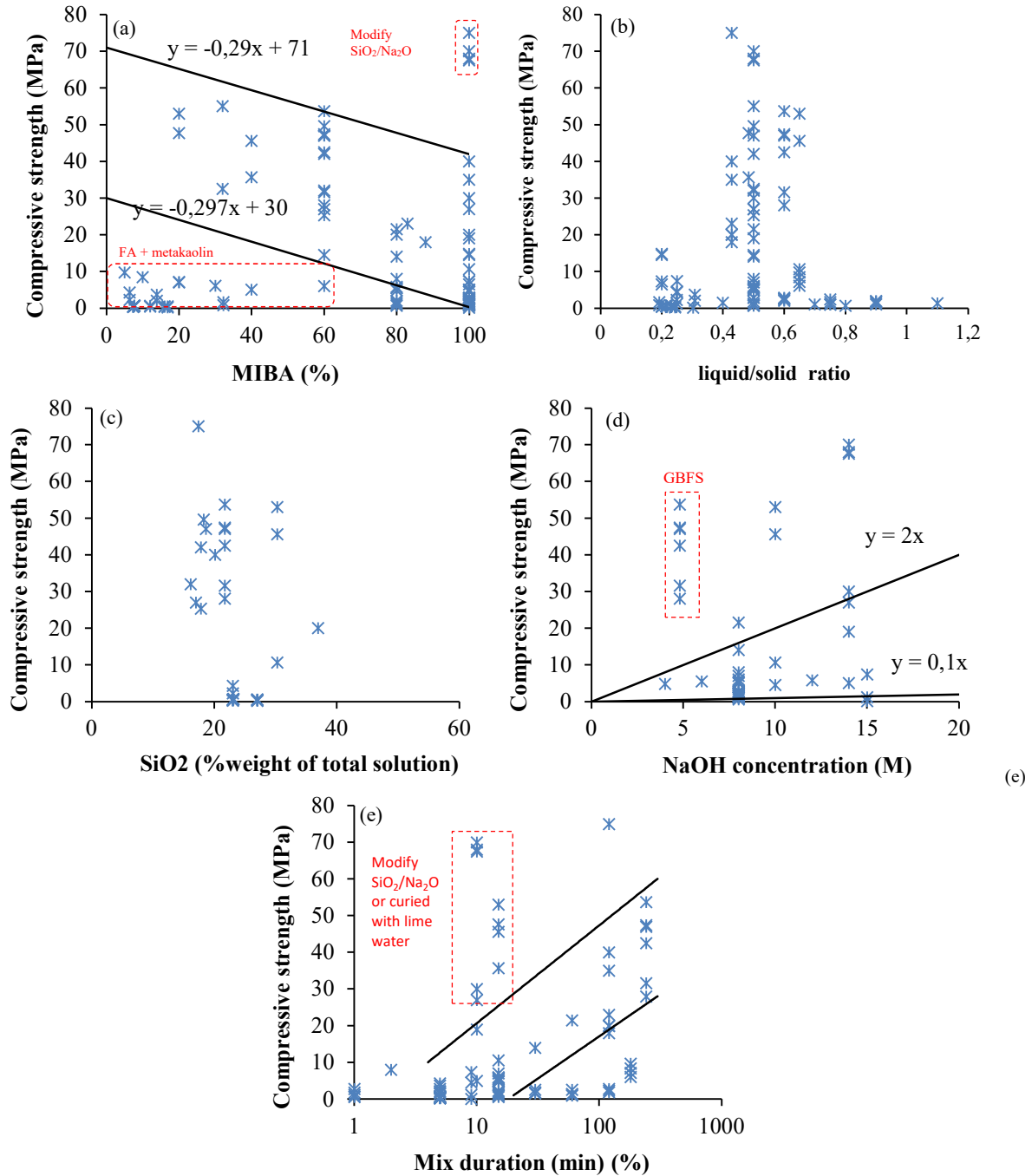


Figure 3 - Compressive strength vs. (a) MIBA incorporation ratio, (b) liquid/solid ratio, (c) total SiO_2 quantity in solution, (d) NaOH concentration and (e) mix duration

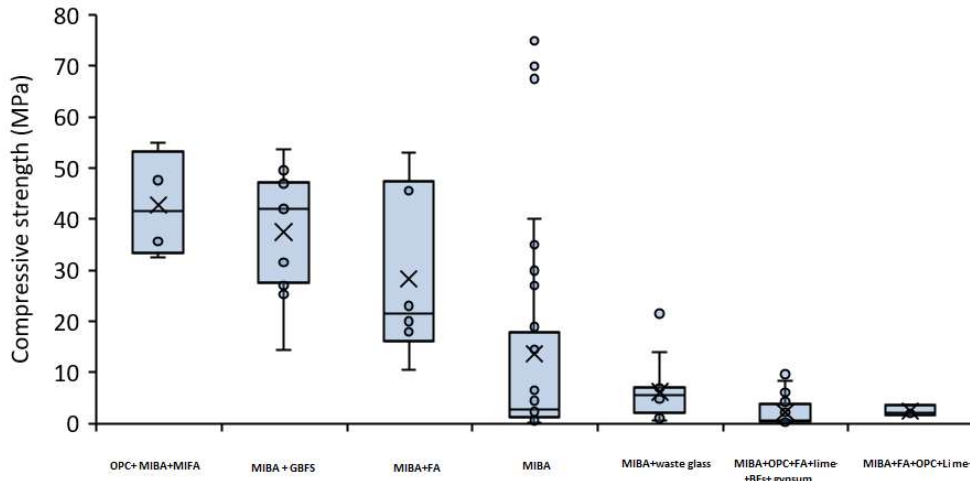


Figure 4 - Effect of various types of precursor on AAM

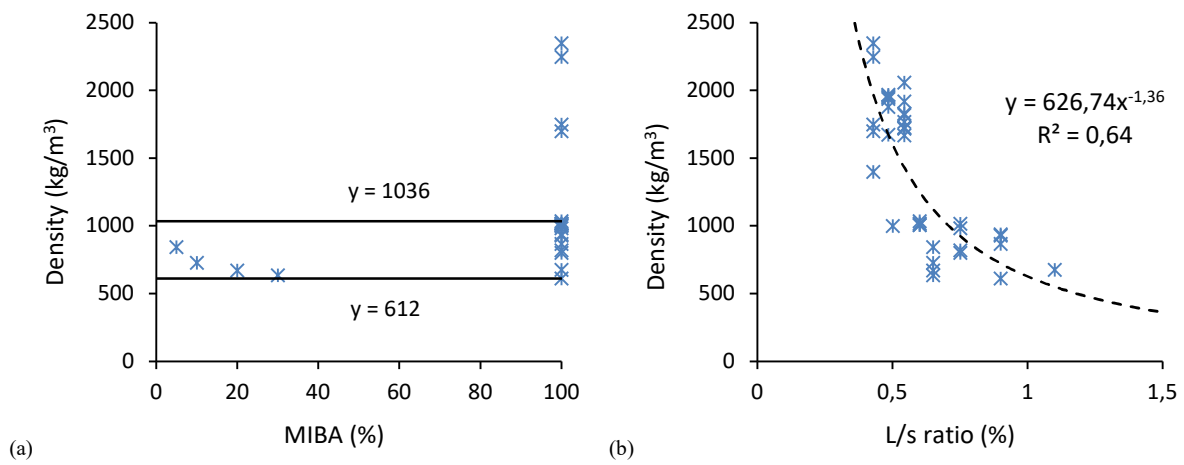


Figure 5 - Density of AAM vs. (a) MIBA incorporation ratio and (b) liquid to solid ratio

The results of Figure 6a show that there is a strong relationship between the compressive strength and density of MIBA ($R^2 \approx 0.83$). Contrary to expectations, no strong correlation between density and porosity was found (Figure 6b).

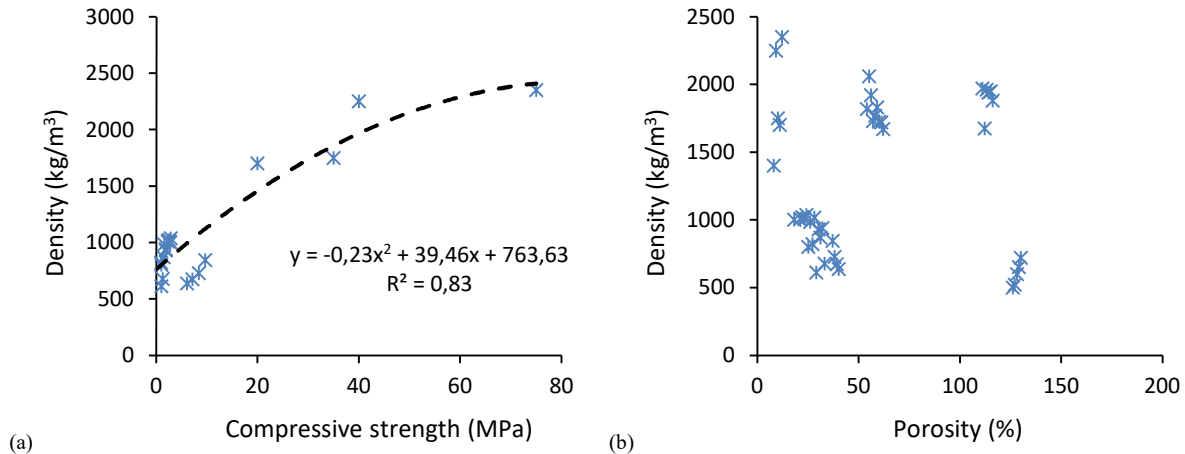


Figure 6 - Relationship between density and (a) compressive strength and (b) porosity of AAM

4. Final remarks

The main objective of this report was to show how the database was built, namely how was the literature review made and which were the selected criteria. The report also shows how some of the missing parameters, which are key for understanding specific behaviours, were determined. According to the preliminary analyses made to the main parameters, it was ascertained that alkali-activated MIBA is a very new research topic with little amount of information and with high potential for development. There are several factors that affect the results and need to be considered, i.e. the type and quantity of precursor, alkali activation and other materials, the curing temperature, liquid to solid ratio, among others. For instance, MIBA-based AAM can be produced with a wide range of compressive strength (0.3-75 MPa) regardless to the incorporation ratio of MIBA. Additional research should be carried out to fill in research gaps and allow a meta-analysis of the results to identify specific trends.

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