DOI
https://doi.org/
10.18690/um.fkkt.1.2025.3

ISBN
978-961-286-959-5

CIRCULARITY AND LONGEVITY OF ALKALI-ACTIVATED MATERIALS: CASE-STUDY USING ROCK WOOL AS A PRECURSOR

BARBARA HORVAT,[1] SARA TOMINC[2]

[1] Milan Vidmar Electric Power Research Institute, Ljubljana, Slovenia
barbara.horvat@eimv.si
[2] Slovenian National Building and Civil Engineering Institute, Ljubljana, Slovenia
sara.tominc@zag.si

Alkali-activated materials (AAMs) are being envisaged as a future alternative to cements because AAMs (i) can be made solely from locally available secondary raw materials and (ii) because of their low energy consumption during synthesis. However, products that cannot be reused should never enter the market. Therefore, the circular economy viability of AAMs was studied by reusing (alkali-activated) pulverised rock wool in alkali activation synthesis until the theoretically determined limit leading to the efflorescence would be reached. While 25% of the maximal allowed liquid alkali did not offer significant mechanical performance, 50, 75 and 100% of allowed alkali ended up with 15, 20 and 20 MPa, respectively, which is only 5 MPa lower than AAM made with the same technique (pressing) in a one-step approach. Therefore, if the initial AAM does not reach the efflorescence limit, it could still be used as a precursor before it becomes waste.

Keywords:
rock wool,
alkali-activated materials,
microwave irradiation,
electromagnetic
dehydration,
linear model,
circular model,
longevity of AAM

University of Maribor Press

1 Introduction

As is known from the field of plastic material, what cannot be recycled, should not be made ('Plastics and the Circular Economy' 2019), and even what can be recycled, should be reused ('Reusing 10% Will Stop Almost Half of Plastic Waste from Entering the Ocean. Heres' How | World Economic Forum', n.d.) to minimise the impact on the environment. This philosophy should be deeply implemented in all industries, the building sector included. Namely, conventional building and civil engineering materials represent a significant burden on the environment by:

- producing 40% of the global man-caused carbon footprint ('The Industry Creating a Third of the World's Waste', n.d.);
- exploiting raw materials in yearly amounts of two-thirds of Mount Everest ('The Industry Creating a Third of the World's Waste', n.d.); and
- the high energy consumption during their production as needed temperatures exceed 1000 °C ('Cement Production: How Hot Air Becomes Green Energy', n.d.).

To reduce the burden on the environment caused by building materials, alternative materials to cement, such as alkali-activated materials (AAMs), are being researched (Škvára 2007; Pacheco-Torgal, Castro-Gomes, and Jalali 2008a). AAMs are presented as an environmentally friendly and sustainable alternative (Obonyo et al. 2014) because:

- they require lower temperatures for their production (below 100 °C; (Pacheco-Torgal, Castro-Gomes, and Jalali 2008b));
- can be made solely from (local) secondary raw materials (Horvat and Ducman 2019);
- show good chemical resistance, i.e., are durable (Bernal, Mejía De Gutierrez, and Rodríguez 1969).

However, AAMs have several issues:

- Waste materials can contain heavy metals and other hazardous species which might not get immobilized in AAM (Sun et al. 2024).

- If the molar ratio of amorphous elements of alkali metal to amorphous Al is not equal or below 1, efflorescence will occur in the AAM and cause deterioration of the product (Horvat and Ducman 2019).
- If alkali earth metals also dissolve and interact with the aluminosilicate network (ASN; also called matrix, gel or binder), efflorescence can get additionally enhanced (if the pre-calculation of the optimal mixture is only related to alkali metal elements) (Horvat and Ducman 2019).
- If from precursor is "used" already everything that could form the ASN in AAM, AAM after its end-of-life cannot be used again as a precursor in alkali-activated synthesis but can be used only as an aggregate or go into a potential acidic reaction chain.

Therefore, if AAMs are made from chemically hazardous materials, the problematic part of the precursor should become completely immobilized in the AAM (Sun et al. 2024). One of the safe choices of the precursor is rock wool (RW) (Horvat et al. 2018; Yap et al. 2021) without organic resin on its surface (Li et al. 2021). Namely, if in alkali activation synthesis RW is without organic resin on its surface, there will be no colouration of the water when AAM from RW is immersed in it and less to no leaching issues. Otherwise, RW should not be used as the sole precursor for the synthesis of AAMs, even though RW is a perfect material for AAMs from the mineralogical and chemical point of view. This is because RW is highly amorphous, has sufficient Si and Al, and a low molar amount of chemical elements from the 1st group of the periodic table, but a high proportion of elements from the 2nd group (Horvat et al. 2018). However, the electron microscopy research confirmed that the ASN has a lower presence of elements from the 2nd group of the periodic table, which means that the elements from the 2nd group do not dissolve in the alkali medium so easily (Pavlin et al. 2021), as is shown in the Supplementary material in Figure S1. Therefore, the significant amounts of the elements from the 2nd group of the periodic table can be excluded from the calculation of the limit value of the alkali addition in the preliminary research. Hence, there is a span left for the addition of alkali to RW while trying to avoid efflorescence.

The theoretical calculation (Horvat and Ducman 2019) of the optimal mixture of the selected precursor (or a set of precursors) and the selected alkali takes into account the results from:

- Loss on ignition (LOI) of the precursor at 550 °C, with which mass percentage of the organic compound is measured gravimetrically.
- X-ray fluorescence (XRF), which determines the presence of the chemical elements (which and how much) regardless of how and to what they are bound.
- X-ray powder diffraction (XRD), which provides a fingerprint of the minerals present in the sample. Because it is unlikely that minerals play a role in alkali activation synthesis, their quantity must at least be estimated. Therefore, the XRD of the potential precursor must be measured together with the standard (internal or external) so that a Rietveld refinement analysis can be performed.

The elemental composition of the amorphous content present in the inorganic part of the sample is then calculated as the difference between XRF and XRD, which is used as the best approximation of the ideal mixture. To achieve the highest compressive strength of AAMs, the molar ratio of amorphous Si to amorphous Al is aimed to be 1.9 (Duxson et al. 2005), and to ensure the lifetime integrity of the material by avoiding efflorescence, the molar ratio of amorphous chemical elements from the 1st group of the periodic table and of amorphous Al must be ≤ 1 (considering that chemical elements from the 2nd group of the periodic table do not contribute to the alkali reaction).

However, even if the precursor used for AAMs does not lead to issues, even if efflorescence was avoided by pre-calculating the mixture, sustainability research on AAMs lacks data. While there is data on the durability of AAMs when exposed to different conditions, there is no data on the longevity of AAMs that exists for concrete (Roman concrete lasts more than 1000 years (Dean 2017), modern concrete 50 to 100 years ('The Problem with Reinforced Concrete', n.d.)). Therefore, this preliminary study focuses on the reuse of alkali-activated pulverised rock wool (RW) in the alkali-activated synthesis up to the mixture that would theoretically lead to efflorescence. AAMs synthesised in a 4-step (circular) model were compared with a 1-step (linear) model in terms of mechanical properties, as compressive strength is the most essential metric in load-bearing building industry products. While the 1st step was below the viability of the material, the AAM prepared in subsequent steps reached 15 to 20 MPa, which corresponds to 60 to 80% of the compressive strength of the linear model. Therefore, this study demonstrates that properly designed AAMs can have extended longevity in the field itself.

2 Experimental

2.1 Materials and characterisation of the materials

The following ingredients were used in the synthesis of AAMs:

- Non-waste RW without organic material (LOI at 550 °C is ~0) on the surface, which was used as a precursor. RW was milled in a vibrating disk mill (Labor-Scheibenschwingmühle TS.250, Siebtechnik GmbH) and sieved below 63 μm;
- Na-silicate solution (Geosil, 344/7, Woelner, 16.9 $m_{\%}$ Na_2O, 27.5 $m_{\%}$ SiO_2, 55.6 H_2O), which was used as an aqueous liquid alkali without further manipulation.

In the present study, the limit for the addition of alkali was set to avoid efflorescence, i.e. the elements of the 1st group of the periodic table (originating from the alkali and the amorphous part of the RW) were not allowed to exceed the molar mass of the amorphous Al (present in the amorphous part of the RW). Therefore, RW (dried, milled and sieved below 125 μm) was analysed by XRF (Thermo Scientific ARL Perform'X Sequential XRF) to determine the chemical composition (elements from fluorine to americium) of RW, and by XRD (Empyrean PANalytical X-ray Diffractometer, Cu X-Ray source; under clean room conditions in the 2θ range from 4 to 70° and step 0.0263°) to determine the minerals present in RW. However, the Rietveld refinement was performed using X'Pert Highscore plus 4.1 software and an external standard (corundum, Al_2O_3) to evaluate the amount of elements present in the crystalline content. The amorphous content is then calculated as the difference between XRF and XRD per element (not per oxide). The results of the RW evaluation are presented in Table 1 and show that RW is completely amorphous.

The theoretically determined mass ratio (using software designed in MS Excel platform, developed in project no. C3330-17-529032 "Raziskovalci-2.0-ZAG-529032" (Horvat and Ducman 2019) and upgraded in ARIS project under grant no. J2-3035) between RW and liquid alkali was 1:0.5.

Table 1: Mass percentage ($m_{\%}$) of crucial elements in alkali activation.

Elements [$m_{\%}$]	Na	K	Mg	Ca	Al	Si	Content [$m_{\%}$]
XRF	1.63	0.34	11.66	16.90	18.61	42.39	100
XRD	0	0	0	0	0	0	0
Amorphous	1.63	0.34	11.66	16.90	18.61	42.39	100

The compressive strength, the most crucial parameter for the load-bearing materials, was measured with the compressive and bending strength testing machine (ToniTechnik ToniNORM) on 1-day-old AAM cylinders.

2.2 Synthesis of the material

The AAMs were prepared in two life-cycle models, i.e. linear and circular (Table 2 and Figure 1):

- The linear life-cycle model (L) was performed as a one-step synthesis (as a reference) where the optimal ratio of alkali to precursor, which should not be exceeded, was calculated using XRF and XRD on rock wool. The mass ratio of rock wool to alkali was 1 to 0.5, and this ratio was defined as the limiting value for the alkali.
- The circular life-cycle model (C) was simulated as a cyclic-stepwise synthesis, where in each cycle 25 $m_{\%}$ of the limiting value of the alkali was added to the (1st cycle: α) rock wool and in each next cycle (50 $m_{\%}$ β, 75 $m_{\%}$ γ, 100 $m_{\%}$ δ) to the microwave-dried and milled AAM from the previous cycle until the sum of the added alkali was equal to the alkali used in the linear model.

Table 2: Chemical composition of the samples (precursor and AAMs) with mixture recipes.

Sample	1st/Al [mol/mol]	Si/Al [mol/mol]	Precursor	Precursor [g]	Alkali [g]	H_2O in alkali [g]	Na in alkali [g]
RW	0.21	1.94	/	/	/	/	/
C_α	0.40	2.09	RW	100	12.5	6.9	1.6
C_β	0.59	2.25	C_α	105.6	12.5	6.9	1.6
C_γ	0.77	2.41	C_β	111.1	12.5	6.9	1.6
C_δ	0.96	2.56	C_γ	116.7	12.5	6.9	1.6
L	0.96	2.56	RW	100	50	27.8	6.3

RW and alkali were mixed at up to 1000 rpm. 25 g of the freshly prepared homogeneous mixture was pressed in a cylindrical metal mould with a diameter of 3 cm and a force of 10 kN. The cylinders were cured for 1 day at 70 °C.

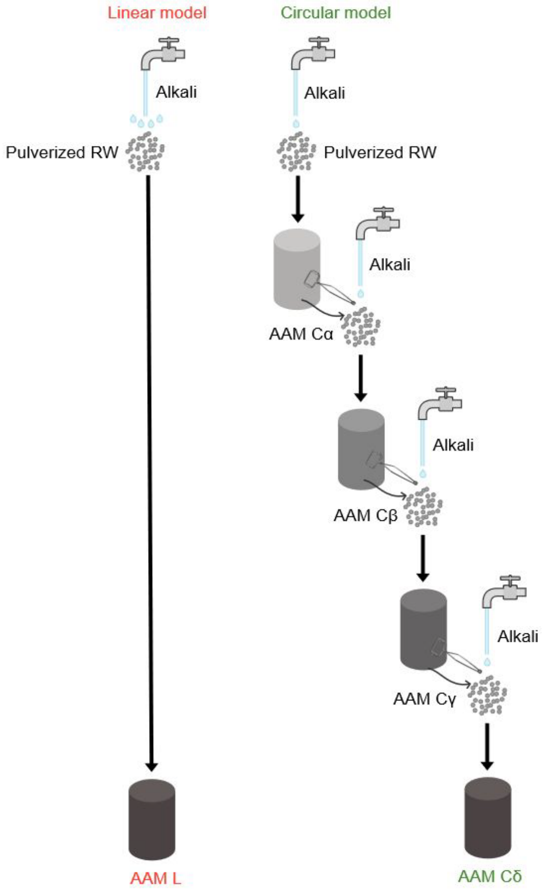

Figure 1: Synthesis scheme.
Source: own.

2.3 Evaluation of longevity of the AAM

The longevity of AAMs, i.e. the lifespan of AAMs during which they can be reused in alkali activation without theoretically causing efflorescence, with the compressive strength of more than 5 MPa (lowest concrete grade available on the market ('Different Types of Concrete Grades and Their Uses | Base Concrete' 2023)), was compared between the two life-cycle models (linear and circular) in two implementation models (design-used and purpose-used):

- Design-used implementation model: All products were used for the purpose they can withstand for time a, after which they were replaced.
- Purpose-used implementation model: All products have been used for the same purpose. Therefore, their lifespan can be assessed on the basis of their physical properties. Assuming that the durability and conditions are the same for all products, the longevity is only influenced by the compressive strength. Therefore, all products were replaced after time $\frac{\sigma_{CS}}{\sigma_1} \cdot a$, where σ_{CS} is the compressive strength of the AAM (in MPa), σ_1 is 1 MPa (to make the measured compressive strength dimensionless), and a is the time.

After the first replacement, the product from the linear model went either to the grave or to another recycling route, while the product in the circular model was reused as the precursor in alkali activation until the total amount of alkali was the same as in the linear model.

3 Results and discussion

All freshly prepared samples in a circular model were sufficiently dry, which means that no liquid was squeezed out of the cylinders by pressing (Figure 2a). In contrast, the samples prepared in a linear model were visibly inhomogeneous (Figure 2b). Inhomogeneity results in parts that have an excessive amount of alkali, where efflorescence forms, and parts that do not have enough alkaline reagent and liquid for complete dissolution of the amorphous content and for diffusion of the dissolved building blocks that rearrange into the ASN of the AAM during dehydration and polymerisation.

The compressive strength (Table 2 and Figure 3) of the samples prepared according to the circular model did not reach the compressive strength of the sample prepared according to the linear model, although they were inhomogeneous (imperfect) immediately after pressing. Because inhomogeneity can never affect the final mechanical properties with an increased value, the compressive strength of the sample prepared in the linear model may not have reached its full potential.

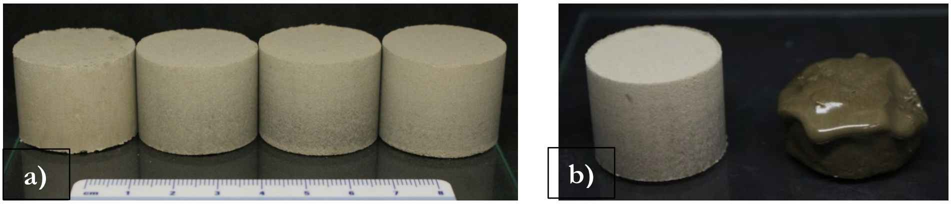

Figure 2: Pressed, freshly prepared mixtures of a) from left to right C_α, C_β, C_γ, C_δ, and b) from left to right C_δ and L.

Source: own.

Table 2: Compressive strength and standard deviation of cylindrical 1-day-old samples.

Sample	Compressive strength [MPa]	Standard deviation [MPa]	Concrete strength
C_α	1.66	1.05	/
C_β	15.94	1.32	Low
C_γ	21.10	2.66	Medium
C_δ	20.45	0.93	Medium
L	27.31	2.51	High

However, the samples prepared in the 2nd to 4th step (C_β, C_γ, C_δ) reached 60% (C_β) and almost 80% (C_γ, C_δ) of the 27 MPa of the linear model sample, which when compared to the concrete can be used for ('Strong Foundations: Mix The Perfect Concrete Every Time' 2013):

- Low-strength concrete (C_β): non-reinforced house foundations, boundary walls, freestanding retaining walls.
- Medium-strength concrete (C_γ, C_δ): reinforced foundations, footpaths, driveways, patios, steps, driveways.
- High-strength concrete (L): suspended structural beams, heavy-duty floors.

While aiming to have the compressive strength at least equal to the lowest value of concrete (5 MPa ('Different Types of Concrete Grades and Their Uses | Base Concrete' 2023)), the circular production of AAMs from RW (cradle) requires skipping C_α directly and supplying the market with C_β first, which should be reused for the production of C_γ, which should be reused for the production of C_δ, before the need to search for an alternative solution to grave: recycling as aggregates in other binding materials in the building industry, as crushed stone-like materials for fences, as alkaline reagents that react with acids and end up in acid-activated materials that do not yet exist, etc.

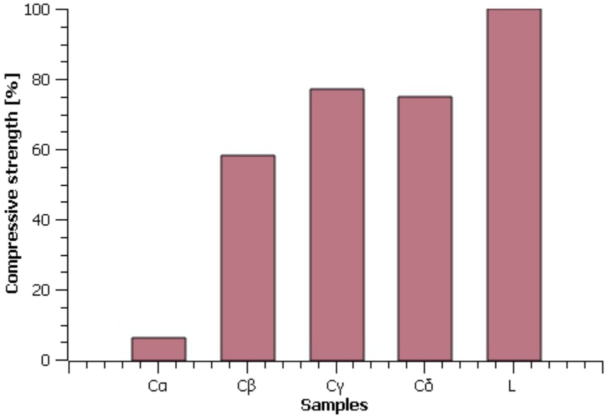

Figure 3: Compressive strength normalised to the compressive strength of the sample from the linear model.

Source: own.

The comparison of the implementation models (design-used and purpose-used) for both life-cycle models (linear and circular), shown in Figure 4, shows the superiority of the circular model. If the product is used as intended (designed), the longevity of the product in the circular model is 3 times higher than that of the material in the linear model. However, if the product is used for a selected purpose regardless of its design (potential), the longevity of the product in the circular model is only 2.1 times higher.

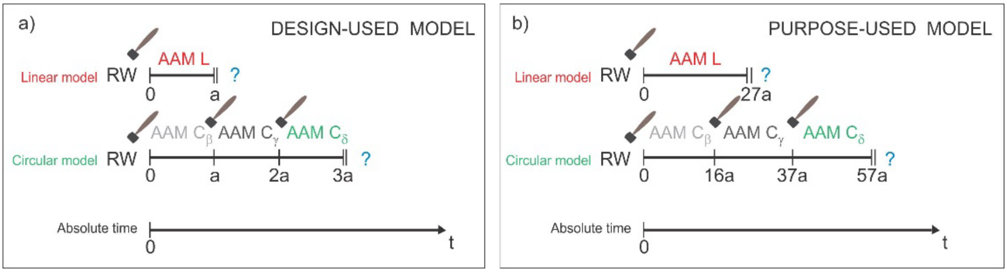

Figure 4: Longevity of the product with marked manufacturing events (hammer), grave or other recycling path (blue waymark) and the current AAM material in the timeline.

Source: own.

4 Conclusion

In this study, the potential of AAM from RW for reuse in alkali-activated synthesis was investigated by assessing the compressive strength and longevity. If efflorescence is avoided in all cases and no other issues occur, the circular model starting at 50% of the maximum allowable alkali extends the use phase of AAM beyond the linear model, regardless of whether the material was used for its intended purpose or not.

Adding 50% of the theoretical limit value of alkali can be compared to low-strength concrete, while adding 75% of the theoretical alkali maximum to medium-strength concrete. However, AAMs in this study were not performed on a pilot product scale comparable to concrete test samples, and concrete cannot be prepared in sample sizes in this study and still be representative.

The fresh alkali-activated slurry can only be pressed until the samples are homogeneous. When liquid is squeezed out from the tablet, slurry has to be moulded and not pressed, just like the liquid can be added to the pulverised precursor up to the point, when buoyancy is equal to forces acting against it in the moulded slurry.

Acknowledgements

This study is part of Dr. Barbara Horvat's ARIS project and was financially supported by the Slovenian Research Agency under Grant No. J2-3035. The present study was performed at the Slovenian National Building and Civil Engineering Institute while Dr Horvat was still employed there, for which Dr. Horvat is thankful. Dr Horvat also thanks the Milan Vidmar Electric Power Research Institute for offering peer review, proofreading, and evaluation of the work by the committee.

Supplementary materials

A polished, uncoated sample of AAM based on rock wool was evaluated using a scanning electron microscope (SEM) and energy-dispersive X-ray spectroscopy (EDXS) under low vacuum conditions (Figure S1) to determine the elemental composition of the ASN (the darkest shade of grey in Figure S1). EDXS mapping showed that Na and Ca were separately concentrated in the unreacted rock wool fibres, that the presence of Na in the ASN was significant (can be attributed to the Na-based alkali used in the reaction), while Ca was notably scarce.

However, regardless of the dissolution rate of Na and Ca present in the rock wool, the lack of Ca in the ASN supports the assumption that excluding elements from the 2nd group of the periodic table in the theoretical calculation is valid (in 1st approximation).

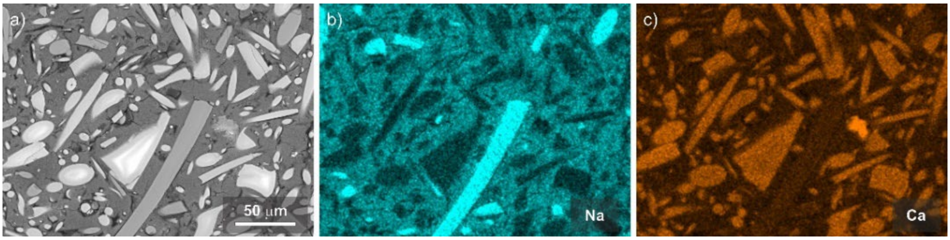

Figure S1: a) SEM micrograph of polished AAM sample based on mineral wool, with EDXS mapping of b) Na and c) Ca.
Source: own.

References

Bernal, Susan A., Ruby Mejía De Gutierrez, and Erich D. Rodríguez. 1969. 'Alkali-Activated Materials: Cementing a Sustainable Future'. *INGENIERÍA Y COMPETITIVIDAD* 15 (2). https://doi.org/10.25100/iyc.v15i2.2608.

'Cement Production: How Hot Air Becomes Green Energy'. n.d. Accessed 30 September 2024. https://insights.thyssenkrupp-polysius.com/story/cement-production-how-hot-air-becomes-green-energy/.

Dean, Amy M. 2017. 'Scientists Research as to Why Roman Concrete Lasts Millennia, While Modern-Day Concrete Lasts a Half Century'. *International Society for Concrete Pavements* (blog). 12 July 2017. https://www.concretepavements.org/2017/07/11/scientists-research-as-to-why-roman-concrete-lasts-millennia-while-modern-day-concrete-lasts-a-half-century/.

'Different Types of Concrete Grades and Their Uses | Base Concrete'. 2023. 1 September 2023. https://www.baseconcrete.co.uk/different-types-of-concrete-grades-and-their-uses/.

Duxson, Peter, John L. Provis, Grant C. Lukey, Seth W. Mallicoat, Waltraud M. Kriven, and Jannie S.J. van Deventer. 2005. 'Understanding the Relationship between Geopolymer Composition, Microstructure and Mechanical Properties'. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 269 (1–3): 47–58. https://doi.org/10.1016/j.colsurfa.2005.06.060.

Horvat, Barbara, Mark Češnovar, Alenka Pavlin, and Vilma Ducman. 2018. 'Upcycling with Alkali Activation Technology'. In *Technologies & Business Models for Circular Economy*. Slovenia.

Horvat, Barbara, and Vilma Ducman. 2019. 'Potential of Green Ceramics Waste for Alkali Activated Foams'. *Materials* 12 (21): 3563. https://doi.org/10.3390/ma12213563.

Li, Zhenming, Irving Alfredo Flores Beltran, Yun Chen, Branko Šavija, and Guang Ye. 2021. 'Early-Age Properties of Alkali-Activated Slag and Glass Wool Paste'. *Construction and Building Materials* 291 (July):123326. https://doi.org/10.1016/j.conbuildmat.2021.123326.

Obonyo, Esther, Elie Kamseu, Patrick Lemougna, Arlin Tchamba, Uphie Melo, and Cristina Leonelli. 2014. 'A Sustainable Approach for the Geopolymerization of Natural Iron-Rich Aluminosilicate Materials'. *Sustainability* 6 (9): 5535–53. https://doi.org/10.3390/su6095535.

Pacheco-Torgal, Fernando, João Castro-Gomes, and Said Jalali. 2008a. 'Alkali-Activated Binders: A Review Part 1. Historical Background, Terminology, Reaction Mechanisms and Hydration Products'. *Construction and Building Materials* 22 (7): 1305–14. https://doi.org/10.1016/j.conbuildmat.2007.10.015.

———. 2008b. 'Alkali-Activated Binders: A Review. Part 2. About Materials and Binders Manufacture'. *Construction and Building Materials* 22 (7): 1315–22. https://doi.org/10.1016/j.conbuildmat.2007.03.019.

Pavlin, Majda, Barbara Horvat, Ana Frankovič, and Vilma Ducman. 2021. 'Mechanical, Microstructural and Mineralogical Evaluation of Alkali-Activated Waste Glass and Stone Wool'. *Ceramics International* 47 (11): 15102–13. https://doi.org/10.1016/j.ceramint.2021.02.068.

'Plastics and the Circular Economy'. 2019. 16 September 2019. https://www.ellenmacarthurfoundation.org/plastics-and-the-circular-economy-deep-dive.

'Reusing 10% Will Stop Almost Half of Plastic Waste from Entering the Ocean. Heres' How | World Economic Forum'. n.d. Accessed 11 November 2024. https://www.weforum.org/stories/2021/07/reusing-plastic-waste-pollution-economy-value/.

Škvára, František. 2007. 'Alkali Activated Material - Geopolymer'. *Department of Glass and Ceramics Faculty of Chemical Technology*, 16.

'Strong Foundations: Mix The Perfect Concrete Every Time'. 2013. Sans 10400 - Building Regulations. 13 March 2013. https://sans10400.org.za/concrete-mixes/.

Sun, Zengqing, Xiaoyu Li, Min Gan, Zhiyun Ji, Xiaohui Fan, and Jinxin Xing. 2024. 'Converting Municipal Solid Waste Incineration Fly Ash and Municipal Sludge into Environmentally Compatible Alkali-Activated Material'. *Sustainability* 16 (18): 7912. https://doi.org/10.3390/su16187912.

'The Industry Creating a Third of the World's Waste'. n.d. Accessed 30 September 2024. https://www.bbc.com/future/article/20211215-the-buildings-made-from-rubbish.

'The Problem with Reinforced Concrete'. n.d. Accessed 1 October 2024. https://www.unsw.edu.au/newsroom/news/2016/06/the-problem-with-reinforced-concrete0.

Yap, Zhen Shyong, Nur Hafizah A. Khalid, Zaiton Haron, Azman Mohamed, Mahmood Md Tahir, Saloma Hasyim, and Anis Saggaff. 2021. 'Waste Mineral Wool and Its Opportunities—A Review'. *Materials* 14 (19): 5777. https://doi.org/10.3390/ma14195777.