Part 1: Background and industry overview

Alkali activated materials – a new generation of cementless binders for concrete

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Concrete structures are widely used for buildings, transportation, infrastructure, and maritime applications. Their design, durability and performance directly influence social and economic growth. The downside of using concrete is linked to issues of durability and enormous environmental costs, as the cement industry is responsible for 8% of the world's anthropogenic carbon dioxide emissions. In addition, around 60% of all non-renewable resources are used in construction, making it one of the least sustainable industries. There is a clear demand for a new, sustainable generation of building materials, as concrete based on Ordinary Portland Cement (OPC) cannot meet all the challenges of modern society in terms of durability and sustainability. The DuRSAAM action addresses this by establishing a training and research network that contributes to a sustainable built environment and uses alkali-activatable materials as a new generation of cement-free binders for concrete.

The design, durability and performance of structures play a crucial role in fostering societal and economic growth. Extraordinary structures can be realised with concrete as building material, and these are often designed for long service lives to gain optimal value from the material, environmental, intellectual and financial input into the making of the structure. Nowadays, the concrete is mostly based on ordinary and blend Portland cement, this is the most used building material in the world and its production is estimated to around 2.6 billion tonnes [1]. This high production of cement represents an important environmental issue, accounting for ~8% of global anthropogenic CO_2 emissions [2, 3]. The productive cycle of clinker generates 0.53 t of CO_2 per ton of clinker produced from decarbonation reaction of limestone and 0.34 t of CO_2 per ton of clinker from combustion of fossil fuel to reach 1450°C. An important contribution to the CO_2 emission, albeit minor, is also given by the grinding and transport phase on which, however, it is more complicated to intervene to reduce the environmental impact.

The rapid growth of global population, especially in urban areas, and the global goals to reduce CO_2 emissions call for alternative solutions that classical cement cannot meet. The cement industries have started to acknowledge the role of alternative binders in a carbon constrained industry, given that there are significant reductions in CO_2 emissions and potential advantages in performance only offered by these alternative binding systems. Alkali-activated materials (AAMs) represent one of these alternative binding systems and, as it will be shown in this paper, a future realistic alternative to the conventional cement responding to the needs for more efficient, durable and sustainable concrete construction.

Alkali-Activated Materials in a nutshell

AAMs is a general name to indicate any binder system derived by the reaction of an alkali metal source (solid or dissolved) with a solid aluminosilicate powder (including geopolymer).

The precursor materials used are a big variety of by-products or waste materials from other industrial activities. Some of them can be used directly, others may need grinding, calcination or thermal activation at 700–900 °C. The most common precursor materials are: slag from iron production, fly ash from coal power plants and metakaolin from calcination of kaolin [4]. In Europe, almost all these materials are already applied as a mineral addition to clinker according to standards and regulations, such as EN 15167-1:2006 for ground granulated blast furnace slag or EN 450-1:2012 for fly ash for concrete. However, these materials are still used only in small fractions as cement substitution. In the case of AAMs, these materials are fully exploited, and even furthermore a big range of other possible precursors are being investigated worldwide, e.g. rice husk ash, slag from copper and zinc, red mud, waste from the production of ferronickel, phosphorous slag, volcanic and synthetic glass, zeolite, general calcinated clay, bottom ash and municipal solid waste incineration [4, 5].

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The activator is second main component of AAMs. Its role is to enable early-age strength development and boost the pozzolanic reactivity of aluminosilicate powder. For AAMs, the activators are based on an alkaline metal, Na or K, and an anionic group, the most common are hydroxide and silicate, but carbonate and sulphate are also quite common. The activation process can be accelerated by adding small amount of clinker or mineral admixture to the dry powder, reducing dimension of the particles of precursors or using heating. The activator may be a liquid solution of one or more components, which is added to the dry precursors, in this case obtained material is called two-part system. On the other hand, a so called one-part systems involves a solid powder to add to the dry precursors and, then, to mix with water [4, 6].

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AAMs can in general be distinguished into two different categories based on the final phase assemblage:

- Low Ca systems – based on the activation of precursor with low Ca content, such as fly ash or metakaolin, where the main reaction product is a three-dimensional alkali-aluminosilicate hydrate (N-A-S-H) type gel, and
- High Ca systems-based on the activation of precursor with high Ca content, such as slag, where the main reaction product is calcium-aluminosilicate hydrate (C-A-S-H) type gel.

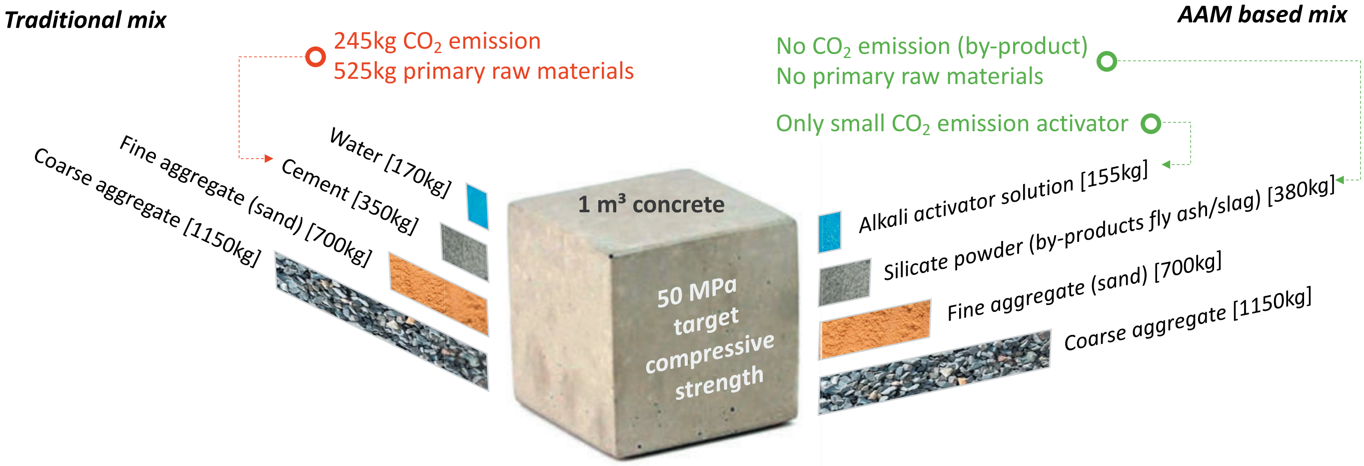
The concept of AAM is illustrated in Figure 1, whereby 1 m³ of the OPC-free AAM concrete is compared with 1 m³ of conventional OPC concrete for a nominal mix composition targeting a concrete compressive strength of 50 MPa. The difference basically comes down to a change in the binder system yet results in profound savings in terms of primary raw materials, CO_2 emission, landfill disposal and costs.

Hereafter, some of the main challenges for wider application of AAMs will be shortly described, together with a proposed action within DuRSAAM project tackling these challenges [7]. DuRSAAM (Durable, Reliable, Sustainable, Alkali -activated material) presents a strong network of 7 academic partners, 13 industrial partners and 2 public organizations focused at underpinning the technological value of AAM as an effective and eco-efficient technology for concrete. Beside this scientific aim, the project is aimed to train a new generation of researchers in the optimal use of AAM concrete for a sustainable built environment, thus creating a critical mass of researchers specifically skilled to address the multi-disciplinary (binder technology, micro- versus macro-structural modelling, transport mechanism and durability, combined environmental actions, fire behaviour, long-term deformations, sustainability assessment) and cross-sectoral (building materials, new concrete construction, structural renovation, durability assessment, eco- efficiency and circularity in industry) challenges.

Challenges for wider application of AAM concrete

Supply chain of raw materials

Since AAMs are based on secondary raw materials and industrial products, their application is directly contributing to the transition towards circular economy and the creation of a real market for these alternative raw materials. However, to ensure a stable and continuous supply of AAMs worldwide, it is crucial to assess the security of future supply of these materials and likely price developments. It is important not to underestimate the risk of unstable supply chain for the main by-products used in AAMs, such as slag and fly ash. A good example is a potential progressive reduction of fly ash availability due to the European announcements of coal phase-out in the electricity sectors. Additional limitation of AAMs based on these common precursors in non-developed countries is the absence of industries producing these by-products. It is therefore of crucial importance to base development of AAMs on locally available mineral residues with a stable supply, which go beyond commonly used precursors.



1m³ AAM concrete:

- ✓ Saves 525 kg raw materials to make cement
- ✓ Saves 245 kg CO_2 emission
- ✓ Uses 680 kg of by-products & recycled aggregates (1)
- ✓ Avoids 70 kg waste disposal (2)
- ✓ Replaces ~ 30€ cement cost (~ 50% of concrete cost) by more cost-effective solution

Targeting 20% concrete market share in EU:

- ✓ Saves 266 Mt/y primary raw materials
- ✓ Saves 10,2 Mt/y CO_2 emission
- ✓ Uses 69 Mt/y by-products & recycled aggregates
- ✓ Avoids 7 Mt/y landfill disposal
- ✓ Cost-effectiveness combined with environmental benefit, targeting a 1B€/y market volume (3)

(1) Assuming 20% replacement by recycled aggregates; (2) Assuming 20% of by-products is currently waste;
(3) 4 tonne concrete per year per capita (EU-28: 512 million habitants in 2017) times 20% share times 30€/m³ and considering 2,4 tonne per m³ concrete

Fig. 1: Comparison between OPC concrete and AAM concrete in terms of constituting materials and potential environmental savings.

Table1: First estimate of availability of relevant secondary raw materials for AAM

Secondary raw material	**Volume (t/y)**	**Remarks**
Common precursors		
Ground granulated blast-furnace slag *Worldwide (estimation)*	ca. 250 million	Heavily utilised in some markets, considered waste in others.
Germany (estimation)	ca. 7 million	5.5 million t/y used for cement
Greece (estimation)	100 000	Partly used as Portland clinker addition
Fly ash *Worldwide (estimation)*	ca. 250 million	Heavily utilised in some markets, considered waste in others
Germany (estimation)	ca. 3 million	High quality material 98% reused in cement and concrete production
Greece (estimation)	10 million	Only a small fraction (< 10%) is sold, moderate quality material.
Alternative precursors for AAM		
EU market		
Ferrous slags	20 million	Not used for concrete production
MSWI ash	15 million	Landfilled or use for road filling
Glass dust	20 million	Remainder of glass collection
Biomass ash	7 million	
Greece		
Rice husk ash	10 000	High quality material
Benelux		
BOF slag	450 000	Iron-containing, less reactive than GGBS
MSWI ash	500 000	Municipal waste incineration ash
Glass dust	14 000	
Slag from secondary copper production	90 000	Iron-silicate based
Bosnia & Herzegovina		
Silica	10 000	High quality material
Electric arc furnace fly ash	800 000	
GGBS	650 000	
BOF slag	150 000	
Bulgaria		
Copper slag	700 000	Iron-silicate fines (ISF)

A rough estimate of the availability of relevant secondary raw materials in the regions of DuRSAAM project partners is presented in Table 1. Extrapolated to Europe and worldwide, this corresponds to a range of millions to billions of tonnes of mineral residues which can be used for cement free binders based on AAM. The table provides the starting point to get an overview of the suitable waste streams currently available. In order to make AAMs competitive with other alternative binders and OPC, they should be adopted to regional availabilities and needs, ensuring postulates of circular economy are fully met.

Conclusions and outlook

Part 2 of this paper will discuss mix composition, fresh and hardened properties, application examples, and aspects related to life cycle analysis for AAM concrete.

AAM are not “one fit all” solutions, rather their main strength lies in the fact that they can be tailored for a specific purpose thanks to the large number of possible combinations from which they can be prepared. At the same time, this adaptability and the numerous possible combinations represent one of the greatest challenges in any generalisation about AAMs and their standardised application in engineering practise. Other challenges include the lack of long-term experience with this novel material, the lack of knowledge about compatible chemical additives and the instability of the raw material supply chain.

The DuRSAAM project addresses some of these challenges by forming a group of 13 early-stage researchers – PhD fellows, each focusing in detail on a specific challenge of the AAMs, but working together to ensure that the knowledge is comprehensive rather than fragmented. The main strength of the project lies in its strong foundation, based on the fundamental knowledge of academic mentors, the experience of industrial mentors and the motivation of early stage researchers.

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