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Life Cycle Assessment of alkali activated materials for pavement applications: preliminary investigation of precursors

Francesca Lolli [1*], Kimberly E. Kurtis [1]

[1] Georgia Institute of Technology, Atlanta GA 30332, USA

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Abstract

According to the OECD database, investments in road networks amounted to €170 billion in 2018, with current practice focused towards portland and asphalt cement concrete pavement. Identifying more sustainable and less energy intensive paving materials is crucial for the expected exponential growth of developing countries. Alkali activated materials concrete (AAM) have been studied with growing interest during the last three decades. AAM show promising results in terms of mechanical performance, while also having a global warming potential impact 30-80% less than that of portland cement concrete. The global warming potential of AAM is closely dependent on the: 1) activating solution used to activate the precursor and 2) the geographical location of the precursor. Specifically, the impact of the transport for both of these components is ~10% of its global warming potential. This study correlates precursor availability, based on global trends in power generation and steel manufacture as well as geological variations, with two key performance metrics: CO_2 intensity and compressive strength. Results indicates that AAM production in the future should utilize MK or other sources of that are locally available in states where the coal industry is being phased out, while in Asian markets, AAM production will likely be based on fly ash and slags. Additionally, AAM display a lower CO_2 intensity when compared to traditional binders, while providing the required compressive strength for pavement applications (30 MPa). Hence, to increase the adoption of AAM for pavements, it is fundamental to identify opportunities for applications that are tailored to the local availability of raw material.

Keywords: Alkali Activated Materials; LCA; Pavements; CO_2 intensity

1 Introduction

1.1 Investments on paved infrastructures in the United States and abroad

The U.S Federal Highway Administration allocates an annual capital investment for construction and maintenance of more than $US 150 billion [1] for more than 2.7 million miles of paved public roads [2]. Future projections for transport-mode scenarios have been hypothesized for the Organisation for Economic Cooperation and Development (OECD) countries [3], on one hand predicting that utilization and construction of roads may decrease in the future with more ridesharing and public transit options, while on the other hand stating that private vehicles will remain the preferred mode of personal travel worldwide. In the near future, investments in road construction and maintenance will likely remain stable, in alignment with the past 10 years trend for OECD countries [3]. Non OECD countries, such as China, have instead experienced a dramatic increase in investments on road construction in the last ten years, reaching over $US 560 billion in 2017 [3]. These planned substantial investments in infrastructure prompt an examination of the current technology, researching alternatives that are able to overcome the intrinsic issues associated with asphalt and portland cement concrete. Specifically the following issues should be addressed:

Environmental impact of the current technology. Both asphalt and portland cement concrete production are associated with an intensive consumption of natural resources while also producing of CO_2 emissions, and water pollution. The extraction from petroleum, often needed for asphalt, and the cement production for concrete causes significant greenhouse gas emissions. In 2017, in the US, asphalt has been responsible for 6% of the consumption of fossil fuel for non-energy use [4], and it should be noted that the production of cement alone accounts for around 8% of the global anthropogenic CO_2 emissions [5].

Frequency of maintenance and repair. Asphalt pavement requires frequent maintenance, resulting in huge investments at the local level [6]. For example the Georgia Department of Transportation invests $US 2-3 billion/year to preserve, reconstruct and maintain road and bridges [6], with more than 95% of the State's roads made with asphalt [7]. However, despite this funding in 2014, only a third of the planned pavement resurfacing was completed, largely due to insufficient funding for resurfacing, which has caused a

* Corresponding author: Francesca Lolli, E-mail: flolli3@gatech.edu

dilation of the maintenance cycle from 15 years to 50 years [8]. To try to reduce the maintenance cycle, in 2015 the State Government allocated an additional by $5.4 billion of investments via the Transportation Funding Act [8].

Furthermore, it should be noted that the existing infrastructure network might not be sufficient to accommodate future increased urbanization. In the next 20 years, like many metropolitan region around the world, the city of Atlanta (GA) is expected to greatly increase in size, reaching a population of 8 million [9] (5.9 million in 2019), straining the lifecycle of the paved network. Even if the strain on the existing and future network will be less than anticipated, due to preference of shared modes of transport over private ones, it will be imperative to improve the ride quality and decrease the rate of repair to assure robustness of the road surface. With the increasing prevalence of autonomous vehicle traffic, along with increased traffic density, reducing the need for repair is of growing importance. It is clear that increasing research in materials with enhanced wear resistance is necessary to provide a longer-term solution. Alternative materials with comparable mechanical properties to asphalt and concrete pavements, but with a more rapid strength development can facilitate rapid construction and large-scale repair and maintenance operations, minimizing road closures and the related environmental and economic costs.

Material costs. The cost of pavement material has risen substantially, even when considering the ~19% inflation rate over the last 20 years, with asphalt costs rising from US$150/ton of 2000 to US$470/ton of January 2020 (US$390/ton July 2020) [10]. It is theorized that alkali activated materials may be produced at lower costs than conventional asphalt or concrete if they are made from by-products of other industrial processes and if locally available materials/wastes are preferred. Furthermore, if combined with a longer service life, additional savings can be added over the lifetime of the infrastructure.

To address some of these issues more sustainable alternatives to asphalt and concrete should be studied for pavements. The following paragraph examines the viability of alkali activated materials as an alternative to current technology, in particular focusing on considerations on their environmental impact. This study will focus specifically on global warming potential of AAM, availability of precursors and corresponding performance of AAM. As a result, recommendations will be made for precursors selection based on CO_2 emissions, their availability and performance.

1.2 Alkali activated cement and concrete

Alkali Activated Materials (AAM) refers to inorganic binders resulting from the activation of an alumino-silicate source with an alkaline solution, typically sodium hydroxide, sodium silicate and/or potassium silicate, with ongoing research on less carbon intensive activators [11]. The final product is a rigid material with comparable mechanical properties to cement-based binders, but with lower global warming potential (GWP) than cement [13-21]. The lower GWP is attributable to the alumino-silicate source used as raw materials or precursors, which are mainly by-products of other industrial processes. Typical examples are: coal fuel ashes, by-product of the coal industry, industrial slags, by-products of steel production, or calcined clays.

To evaluate the environmental impact of AAM as compared to asphalt or concrete production, a Life Cycle Assessment (LCA) approach is recommended [12]. For cement-based materials, LCA follows the “cradle to gate” approach, considering all the components of the production process (cradle) through bringing the product to market (gate), excluding the impacts from transportation, placement, maintenance and disposal of the product throughout its full life cycle. This is a common approach because cement-based materials are used in various end-products, which affects their environmental impacts in a non-informative manner. It should be noted that while transport of the final product is not considered in this approach, transportation of the precursors to the production site should instead be accounted for. A recent publication [22] analyzed the impact of different scenario for FA transportation on the overall GWP of concrete concluding that international import/export might effectively compromise the benefits of using FA as partial replacement for cement. The CO_2 related to the transportation to and from international sources might be higher than the CO_2 gains due to the use FA as SCM. LCA discretizes the impact of the material production into several impact categories, but for the construction sector, the global warming potential (GWP) category is the most important impact, since cement production is a leading contributor to global CO_2 production. When comparing GWP among construction materials, the most conservative practice is to assume equal volume among the replacements.

2 Methodology

Since 2010, over 2000 articles have been published comparing AAM and portland cement binders, but at present, only a small number of peer-reviewed papers have been published on the CO_2 emissions associated with the production of AAM. Robust information is needed for decision-making in the context of sustainable development of infrastructure [11]. Of the relevant papers in this field, 9 provided information on mechanical performance and environmental impact for both AAM and reference cement binders [13-21], and were thus examined in this study. The consensus among the 9 papers is that from LCA studies, AAM have a lower GWP than portland cement, and it appears that the main source of CO_2 impact for AAM is the production of the sodium silicate solution. There are however, aspects of these LCA studies that warrant further consideration. First, while in this study only the GWP impact category is considered as mentioned prior, AAM might display a higher ecotoxicity (ET) and human health impact (HT) when compared to portland cement [17,20].

Second, predicting the impacts of the sodium silicate solutions is hindered by outdated data on the production process [23], based on the environmental performance of 1995 Solvay technology. It should be noted that sodium carbonate, the raw material used to obtain sodium silicate, can either be produced through the Solvay process or directly sourced by mining carbonate salt deposits. Direct mining technology involves 1/10 of the CO_2 production of the Solvay process, hence prior efforts might have likely overestimated environmental impact because alternative sources were not considered [24].

Third, LCA studies may neglect to consider the local availability and composition of the AAM's precursor materials. Due to the abundance of raw materials, and thorough diffusion of cement production technologies, cement production is now an established market with a wide availability of production sites around the world. On the contrary, AAM are not produced extensively, due to a lack of established market infrastructure and the relative concentration of activators, which may result in a large CO_2 impact from transporting the materials to market [25].

Hence, the focus of this work is to analyze and synthesize a selection of published LCA literature [13-21] relating the availability of raw materials with research conducted on AAM. The data collected are specifically referred to:

- Performance parameter expressed as CO_2 intensity
- Location of the conducted research
- Raw materials employed

As a result, recommendations will be made for precursors selection based on CO_2 emissions, their availability and performance, with the aim of tailoring the specific type of AAM to the geographic location.

3 Results

3.1 Worldwide availability of raw materials for AAM production

Three alumino-silicate materials have been identified as the most common precursors for alkali activation: fly ashes (FA), ground granulated blast furnace slags (GGBS), and metakaolin (MK) (calcined kaolin clay). Based on published data [26-30], Fig. 1 shows the availability of these raw materials on a global scale and Fig. 2 summarizes the FA and GGBS availability per continent.

FA are a by-product of the coal industry, hence in Fig. 1 the availability of FA is considered to be proportional to the produced power in MW from coal plants (data from 2018) [26]. In Fig. 1 only power plants with a capacity of more than 30 MW are tracked, and it should be noted that the location of the bubble does not represent the specific location of each power plant, but rather are the geographic center of the respective countries, for location specific data see [26]. The availability of GGBS, a by-product of iron production, is considered to be proportional to their exported value in 2017, with each bubble representing the economic value associated with the trade of GGBS. For GGBS the references used combine trade data of main exporters and importers of GGBS [27], and steel plants locations [28]. MK is the product of the calcination of kaolinite, hence its availability has been represented by considering the worldwide availability of topsoil kaolinite, with data from Ito et al. [29].

Fig. 1 presents an overview of the abundance of key raw materials for AAM worldwide. From Fig.1 the most widely dispersed raw material appears to be kaolinite. Kaolinite is one of the most abundant natural clays worldwide. It is found predominantly in humid areas such as North and South America, Eastern Europe and Russia, Central Africa and Eastern Asia, both in the top and subsoil (easier for extraction the closer to the surface). Metakaolin, the form needed to produce the AAM, is traditionally produced by calcining kaolinite at around 750°C, although higher temperatures may also be used [30]. The current uses of kaolin are mainly in the ceramic and paper industry, and MK as supplementary material for cement-based binders.

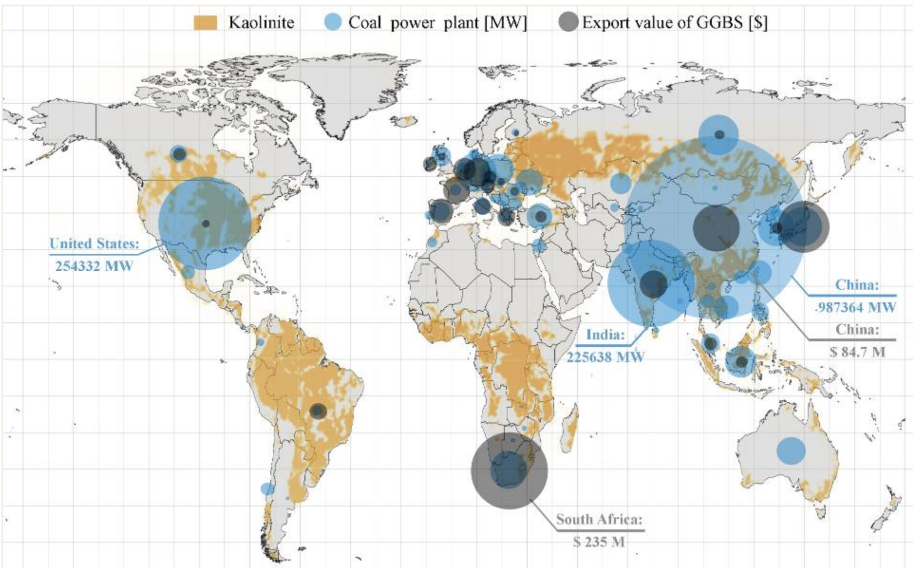

Fig. 1. Map displaying the worldwide availability of the raw materials used for AAM production. For coal power plants, the area of each bubble is proportional to the capacity in MW of the power plant. For GGBS the area of each bubble is proportional to the economic value associated with the export of GGBS.

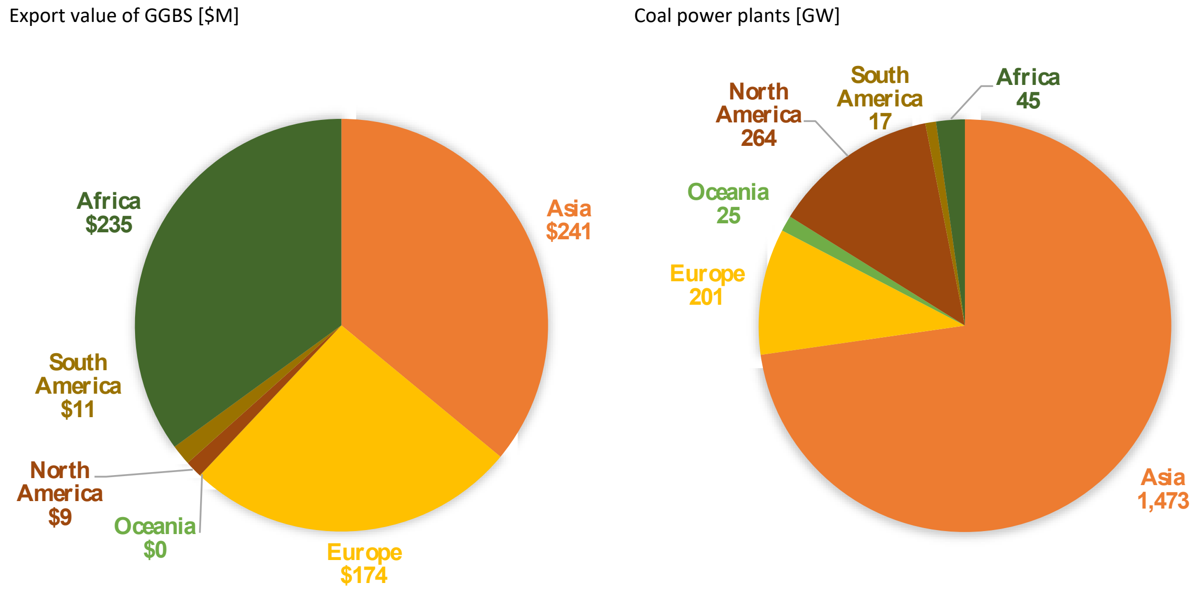

Fig. 2. Pie chart displaying the availability of GGBS and FA per continent (data from Figure 1 summarized by continent).

From Fig.1 it appears that the availability of FA is more geographically dependent, due to its reliance on coal combustion, which is decreasing in the European and US market as renewable and gas fuel electricity production is promoted [26, 31]. In the past, FA have been stored in onsite retention ponds, and reclaiming those deposits as requalified land is attracting significant interest [32-33]. The main challenge is to find uses for this waste material, which is characterized by a wide variability in chemical composition. AAM might be a suitable application for reclaimed FA. The Chinese and Indian market exhibit a reversed trend to Europe and US, with new planned coal power plants to be completed in the next years as a result to their economic growth [26].

GGBS production, which is reliant on iron production, follows a similar trend to that of FA, with the Chinese and Indian market increasing their production/export, while the US and European market are decreasing it. Japan and South Africa remained instead primary exporters in the last 10 years. The decrease of GGBS availability in Europe and the US results from the recycling of steel (a sustainable practice) and a decline in the production of virgin steel.

The overall findings are that MK-based AAM could be produced efficiently worldwide, whereas FA and GGBS based AAM rely on more geographically-dependent resources. Therefore, to reduce the transportation-related emissions, FA- and GGBS-based AAM should be produced more readily in Asia and South Africa.

3.2 Comparing the availability of raw material to the published LCA articles

While acknowledging the limited size of the dataset analyzed, it could still prove worthwhile to compare the geographic distribution of available raw materials in Fig. 1 to the published LCA studies in those areas. Intuitively, AAM production and scientific interest should be correlated to the local availability of precursor materials. This hypothesis is generally observed when examining the 9 LCA studies in Table 1 [13-21]. These studies were chosen since they explicitly considers GWP and mechanical performance of both reference cement binders and AAM.

Table 1. Comparison of published LCA articles.

Location	Raw material	Reference
Australia	FA; GGBS	[16]
Australia	GGBS; FA	[19]
Central America	Zeolite	[13]
Central America	Red Clay Brick Waste	[21]
Central Europe	MK; GGBS; FA	[14]
Italy	FA	[20]
South Korea	MK; GGBS; FA	[15]
United States	GGBS	[18]
Western Europe	MK; GGBS; FA	[17]

As previously anticipated, the most frequently used materials for alkali activation are MK, FA and GGBS with the exception of two recent studies [13, 21] which adopted natural zeolite and red clay brick waste, both materials locally available. A clear exception to the hypothesis is the US study for which GGBS is used as the raw material, even if the current supply is decreasing. Considering the relative abundance of MK in the US, one area of future work might focus more on MK-based AAM, particularly impure MK due to financial considerations (90% less expensive than pure MK [34]). As mentioned prior, another field of research that is likely to receive significant interest is the development of AAM from local reclaimed FA, which may present economic benefits over MK. Countries like Australia, however, have low relative availability of kaolinite, and thus are unlikely to shift to a MK-based AAM, even if the amount of FA decreases in the future from reduced coal utilization for power. Instead, new local materials should be preferred, such as in [13, 21]. Practices such as importing fly ash, even when reclaimed from ponds, should be carefully considered accounting for the embedded CO_2 due to the trasportation to the production site [22].

3.3 CO_2 intensity

To clarify the link between environmental impact and mechanical performance, data from the analyzed LCA literature [13-21] has been represented as a function of CO_2 intensity in Fig. 3. Fig. 3 distinguishes between AA binders and OPC binders, while Fig. 4 presents the AA binder results categorized by precursor used. The main sources for CO_2 for AAM are related to the activators generally composed of sodium hydroxide and sodium silicate. Sodium hydroxide is produced via the chloralkali process, consisting in the electrolysis of sodium chloride to form sodium hydroxide, hydrogen and chlorine. Following the ban on mercury cell technology in 2010 [35], currently the membrane cell process is the preferred technology. Sodium silicate is produced by calcination of silica sand and sodium carbonate at 1000°C. The raw material sodium carbonate can either be sourced through the Solvay process or by mining carbonate salt deposits (with 1/10 of the CO_2 emissions of the Solvay process) [24]. CO_2 intensity defines the amount of embodied CO_2 in a cubic meter of material with a specific compressive strength. As a reference, a typical pavement application requires a compressive strength on the order of 30 MPa (as indicated in red in Fig. 3), while flexural strength and stiffness are commonly used in pavement design, not all studies report those properties, although most do report compressive strength. Relationships between compression strength and flexural strength and stiffness (and even wear resistance) have been established, making this measure a helpful indicator of performance of pavement materials.

Fig. 3 distinguish between three categories of materials: alkali activated binders (both mortars and concrete binders are considered), portland cement binders (both mortars and concrete) and cementitious binders with both portland cement and supplementary cementitious materials (GGBS and FA). Based on the trend lines for all the classified materials, an increase in compressive strength corresponds to a decrease in embodied CO_2. That is, an inverse relationship is found between mechanical performance and CO_2 intensity, for all three binding systems examined. This result is also reported in the literature [36], likely due to more efficient use of clinker at higher strengths. The amount of binder used to produce concrete can be highly variable, allowing for a potential reduction of cement used in the mix. It is generally assumed that lower CO_2 intensity can be achieved by replacing cement with SCMs, however the GWP is highly dependent on the source of SCM and in particular if these are carbon neutral or not. In fact, it has been reported that Portland cements with a clinker factor of 95% can display lower intensity than concretes with a lower amount of clinker [37], reinforcing the conclusion that choosing a local source of SCM is a fundamental step to mitigate the CO_2 intensity of concrete.

From the analysis of these studies traditional portland cement binder and PC and SCM binders display a similar trends, with only a clear environmental advantage of PC+SCM binders for concretes with compressive strength over 40 MPa. This coincides to a large extent, however, with strengths required for this application, making PC+SCM binders preferable from the perspective of minimizing CO_2 emissions while achieving satisfactory mechanical performance.

Alkali activated binders display reduced CO_2 emissions with low correlation to the compressive strength when exceeding ~20 MPa. Between 20 and 70 MPa AAM show a consistent 50% reduction in the CO_2 intensity compared to PC and PC+SCM. In particular at ~40MPa, and ~70 MPa where more data points are available, it can be seen that PC is clearly the most CO_2 intensive binder, followed by PC+SCM and AAM with lower values. The same cannot be clearly observed at ~25 MPa were PC and PC+SCM are overlapping at ~14 kg CO_2/(m^3MPa).

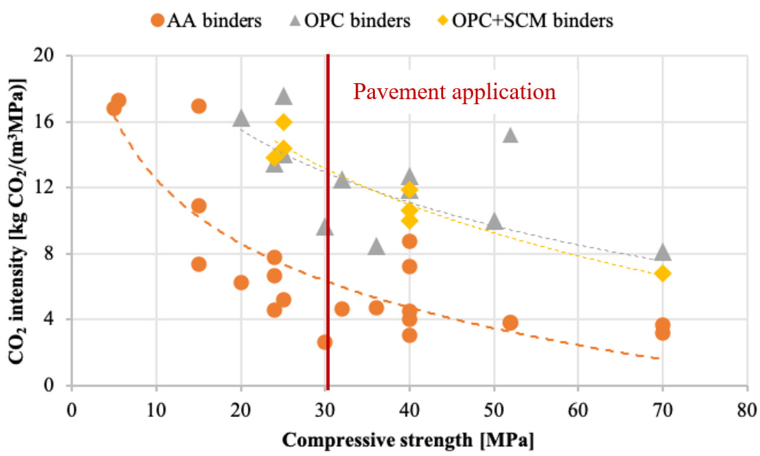

Fig. 3. CO_2 intensity as a function of compressive strength. Data from published LCA literature [13-21].

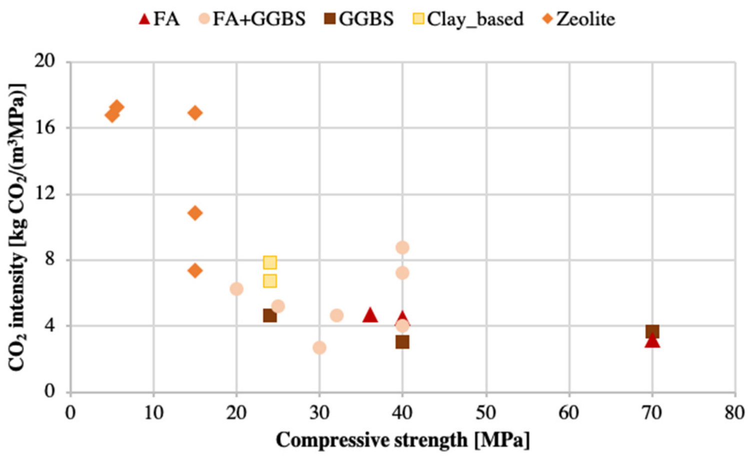

Fig. 4. CO_2 intensity as a function of compressive strength for AA binders distinguishing the precursors used. Data from published LCA literature [13-21].

The variability among the AAMs visible in Fig. 4 is likely correlated to the different precursors and different activating solution used for the mix design. Fig. 4 shows that while FA, GGBS and clay based AAM display similar CO_2 intensities on a wide range of compressive strength, zeolite based AAM are more variable. This is due to the type of activator used for the mix design: the higher CO_2 intensity are correlated to imported activating solution with associated transportation emissions.

When considering pavement applications, Fig. 3 and 4 suggests that AA binders may provide the required strength with considerably less CO_2 emissions than traditional technology, though future work should be performed to ensure that durability indicators (e.g. permeability and wear resistance), are also considered.

4 Conclusions

This is the first attempt to correlate precursor availability, based on global trends in power generation and steel manufacture as well as geological variations, with two key performance metrics: embodied CO_2 and compressive strength. This analysis is based on synthesis of nine published studies, which individually describe these performance metrics for a narrow range of concrete compositions. From examination of these data together, the following conclusions can be made:

- Globally, it is likely that significant resources will continue to be directed toward road construction and maintenance. The spending is likely to stabilize in the OECD countries, and experience a large increase in countries such as China; tolerance for maintenance frequency is likely to decrease, with increased traffic density and increasing autonomous traffic.
- The market for AAM should benefit from the relative abundance of raw materials globally and the stated interest in reducing CO_2 emissions, of which cement production contributes significantly;
- Due to the transition to renewable and alternative sources of electricity, as well as the relative reduction in steel production, it is likely that AAM production in the future will utilize MK or other sources of that are locally available (e.g. reclaimed FA) in particular in the US and European market, where the coal industry is being phased out in many states [25, 29] ;
- Due to the increasing demands for construction materials and energy in the growing Asian markets, AAM production will likely be based on FA and GGBS in those areas;
- From the LCA studies that have been analyzed, AAM display a 50% reduction in CO_2 intensity when compared to PC and PC+SCM binders, while providing the required compressive strength for pavement applications (30 MPa).
- Zeolite based AAM display the higher CO_2 intensity and the lower compressive strength, and therefore are found not to be suitable for pavement applications.

Future research should focus on the design and testing of rigid pavements from AAM. The ultimate goal is to develop guidelines aiming to minimize CO_2 emissions while achieving adequate strength, and acknowledging that different raw materials might be adopted depending on the local availability.

Acknowledgements

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