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Cementitious binders and reclaimed asphalt aggregates for sustainable pavement base layers: potential, challenges and research needs

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Abstract:
Reclaimed asphalt pavement (RAP) has been increasingly used in the past few decades as a replacement of natural aggregates in pavement layers. However, low replacement level is employed, and consequently, large quantities of RAP are still left unused. Researchers often neglect foundation pavement layers, but it has higher prospects to consume recycled materials due to their increased thickness. RAP has inferior properties compared to natural aggregates; therefore, cementitious binders are used to produce a (sub)base layer using high volumes of RAP. This paper reviews the use of RAP aggregates with cementitious materials for pavement foundation layers. Special attention is given to the use of alkali-activated materials (AAM) as a binder in substitution for Portland cement (PC). This review discussed the properties of fresh and hardened RAP in cementitious matrices, as well as changes in the microstructure. The biggest challenge on the use of RAP on both systems, RAP-PC and RAP-AAM, is the bond issues caused by the presence of asphalt on the surface of the aggregates. Some researchers addressed how physical or chemical pre-treatments to the RAP could improve the adherence to the paste, but few studies focused on the optimisation of the binder. A literature survey indicated that an optimised mix design, durability studies and life cycle assessment (LCA) are important research needs towards the development of RAP-AAM. Despite the lack of research evidence, RAP-AAM is a promising solution for foundation pavement layers.

Keywords: RAP; alkali-activation; pavements; base-layers; recycled aggregates

1. Introduction
The road infrastructure comprises 21 million kilometers worldwide [1], and the related industrial sectors are said to be responsible for 24% of the global Greenhouse Gas (GHG) emissions worldwide [2,3]. Natural aggregates are a non-renewable source and a major component of pavements, any small recyclability actions may significantly reduce the environmental impact of the infrastructure sector. Currently, pavement sustainability actions comprise the employment of several types of recycled aggregates. The most used recycled aggregates are recycled concrete [4], steel slag [5], waste foundry sand [6], waste glass [7], crushed brick [8] and reclaimed asphalt pavement (RAP) [9].

RAP is a waste produced during the asphalt road rehabilitation; Europe and USA combined produced well over 100 million tons of RAP in 2017 [10]. In Europe, 68% of the reclaimed asphalt is reused in asphalt mixes, 19% is used as granular materials in unbound layers, 1.25% finds application in other civil engineering projects and 11% ends on landfills [10]. Most studies recommend the use between 20-50% of RAP as an aggregate replacement; in the US, many of the State Department of Transportation (DOT) agencies limit the RAP content to 15% to avoid variabilities in the hot mix asphalt [11]. Zhang et al. [12] presented a detailed review of the production process and performance of RAP-containing asphalt hot mixes. In Belgium, RAP also finds applications in cement-stabilized and loose bases / subbases [13].

Worldwide a relatively high amount of RAP is yet not recycled and therefore disposed of in stockpiles. Other recycling actions are in place, such as the utilisation of RAP as a replacement for natural aggregates in Portland cement (PC)-based materials (mainly concrete). However, the poor adhesion of
RAP granules to the cement paste compromise the mechanical and durability properties of the concrete [14].

Alkali-activated materials (AAM) emerged in the last two decades as sustainable building materials to replace PC for some applications. These new binders rely on the alkaline activation of natural materials (mainly clay and metakaolin) and industrial residues or wastes (such as pulverised fly ash, ground granulated blast furnace slag, mining residues, among others). AAM has been studied as stabilisers in base-subbase layers and more recently employed in combination with RAP aggregates [15,16]. Although the results appear to be promising, the literature on this subject is still scarce. To the author’s knowledge, there is no publication addressing the advantages and shortcomings of alkali-activated materials containing RAP aggregates (RAP-AAM).

Most of the researches on sustainable materials for pavement sections focus on the asphalt layers and undermine the potential of the foundation layers (base and subbase). The foundation layers are thicker than asphalt layers and therefore, can consume higher volumes of recycled materials. However, the use of larger quantities of recycled aggregates represents, in general, a loss of mechanical properties that need to be compensated with the use of cementitious binders or stabilisers. The most used stabiliser for base layers is PC, a material with a high carbon footprint and therefore not sustainable. One of the possible substitutes for PC is AAM. An adequately designed alkali-activated binder could contribute to a reduction on carbon footprint, high early strength, low water permeability and higher resistance to chemical attacks [17,18]. However, designing the alkaline-activated binder is not an easy task, especially when aspiring for sustainability: the type and amount of raw materials must be carefully selected. Miranda et al. [19] performed a test track using AAM for the soil stabilisation of base layers and observed an equivalent mechanical performance to traditional binders (PC and lime). The authors suggested that the mix design for this kind of application must be further optimised and recommended the use of solid activators.

Recently there has been a significant rise in publications investigating construction materials with high RAP content and the use of alternative binders (AAM). The use of RAP-AAM could reduce the environmental impact of the construction industry as well as the costs. The main question to be answered is whether RAP-AAM is a sustainable and suitable material for pavement layers, particularly base layers.

This review focus on the use of RAP as an aggregate replacement in cement bound pavement sections and the use of alkaline binders as alternative cementitious systems. We provide an overview of what has been published in the literature, the existing knowledge gaps and available opportunities. Some of the topics discussed herein are:

(i) How the research of RAP in cement bound pavement layers has evolved over time.
(ii) Which AAM is more suitable for road application.
(iii) Which methodologies are the most used to study RAP-AAM.
(iv) How the performance of RAP-AAM compares with the currently used practice.
(v) How RAP-AAM is related to sustainability

After this introduction, Section 2 presents the state of the art of the subject. It will start with the main properties of RAP, how it is being investigated as an aggregate replacement for pavement layers, and the properties of RAP in PC matrices (fresh, mechanical and durability). The use of alkali-activated binders as PC replacement will also be introduced in Section 2, followed by the main differences between systems with high calcium content and low calcium content, and the extent of the research so far in RAP-AAM. Section 3 debates the literature findings and proposes future research needs, and Section 4 presents the conclusion.

2. State of the art

2.1. Reclaimed asphalt pavement (RAP) aggregates

RAP is a material obtained during the removal of distressed pavement sections for renovation. The EU standard NBN EN 13108-8:2016 defines reclaimed asphalt as ‘the processed site-won asphalt, suitable and ready to be used as a constituent material for asphalt, after being tested, assessed and classified according to this standard’. The assessment includes particle size distribution, type of binder and content, presence of foreign matter (cement concrete, bricks, subbase material, synthetic materials, metal, wood or plastics), as well as the homogeneity and frequency of testing of the feedstock.
Milling machines may be costly and, in some developing countries, backhoes and bulldozers are often used to rip and break the pavement in an uncontrolled milling procedure. Ripping and breaking the pavement brings variations in the characteristics and quality of RAP aggregates [20].

For many years, the production of new (hot and cold) bituminous mixes incorporates some amount of RAP. There is an indication that the replacement of virgin aggregates (VA) by RAP has a positive effect on rutting and stiffness and contradictory reports on the fatigue resistance of pavements [21–26]. Nevertheless, the employment of RAP in surface pavements has become common practice in many European countries such as Germany, Netherlands, Denmark and Belgium [27]. The employment of RAP in new asphalt mixes is beyond the scope of this review. Antunes et al. [9] and Zhang et al. [12] recently published a comprehensive study of this subject.

Despite the recent recycling activities, large quantities of unused RAP are disposed of in stockpiles or landfills worldwide [28]. Recently, it is possible to see an effort to promote a higher use of RAP in unbound layers and other pavement layers (base or subbase). RAP can be used as a substitute for VA, thus reducing its disposal and demand [29]. Some studies claim that the recycling of RAP could reduce the overall cost of pavement sections by 40%-46% [30,31].

The biggest challenge on the use of RAP on cementitious binders is due to the thin, aged bitumen coating on the surface of the aggregate, which leads to inferior properties [32,33]. RAP performance is often improved by blending with VA or stabilising with additives.

In general, RAP aggregates are inferior to VA regarding its gradation, strength and stiffness [32]. The hydrophobic asphalt layer on the interface of the aggregate is responsible for the lower water absorption of the material and the weak bonding between the aggregate and the cementitious mortar [32,34–36]. According to Saride et al. [32], the exposed area of aggregates ranges from 15% to 70% (averaging 34%), but beneficiation process can help on the partial or complete removal of the coated asphalt. Some beneficiation methods use solvents to facilitate the cleansing of the aggregates, but those are considered expensive and unviable at large scale [34].

Singh et al. [34] studied the impact of several RAP beneficiation methods on the mechanical properties of PC concrete produced with treated RAP aggregates presented lower water absorption, coefficient of sorptivity and permeable voids than the ones produced with VA. Brand et al. [37] compared washed and “dirty” coarse RAP replacement (up to 50%) and concluded that washing did not improve the strength of the concrete.

Screening through the 4.75 mm sieve separates the coarse and fine fraction of RAP. Fine RAP is coarser than natural sand aggregates, likely due to the bitumen acting as an adhesive to fine RAP particles [30,38]. Both fractions show lower specific gravity and bulk density than natural aggregates, possibly a consequence of the low-density of the asphalt coating. The aggregates may present a higher water absorption due to the increased water demand from the dust layer [30,31,39].

Age may also play a role in the properties of RAP aggregates. Singh et al. [40] observed that coarse RAP aggregates from new pavements yielded a concrete less workable and with worse hardened properties than an old RAP. The new RAP aggregate was from 2.5 years old pavement freshly milled, while the old aggregate was from 20 years old pavement with eight months of stockpiling. Older RAP aggregates originated from pavements which have been a long time in service or stockpiles have more oxidised asphalt layer. Brand and Roesler [41] suggested that chemical oxidation could successfully modify the surface chemistry of the asphalt leading to better wettability and bonding with cement paste. The authors used an oxidation/ pre-treatment for asphalt: nitric acid (HNO₃), sulfuric acid (H₂SO₄), hydrochloric acid (HCl), phosphoric acid (H₃PO₄), potassium permanganate (KMnO₄), maleic anhydride (C₂H₄(CO)₂O) and ultraviolet light (UV).

2.2. The use of RAP aggregates in pavements base layers

Pavement structure comprises several layers and all of which offers a different opportunity for the use of more sustainable materials. Despite the overall choice of the pavement structure (flexible, semi-rigid or rigid), lower layers present the higher prospects to incorporate high volumes of sustainable materials (especially cement-treated subbase/base layer) [2]. The typical base layers can be unbound granular,
asphalt treated, cement treated, permeable and recycled. Whereas subbase may be absent if there is an appropriately strong subgrade or low traffic [42].

Figure 1 illustrates the typical/possible cross-sections of different types of pavements [2]. Sustainable actions on materials for those pavement layers mostly consider replacing natural aggregates for recycled ones and also replacing PC used as binder/stabiliser with greener binders.

RAP and other recycled aggregates are a suitable alternative when opting for unbound layers (layers GB and GS in Figure 1). However, the most significant opportunity to use a combination of RAP with AAM for pavement layer is while considering cement-treated bases (such as layers CT and maybe GS) and concrete layers (RC, LC and LCL).

![Typical cross-section of pavements](Image)

Figure 1. Typical cross-section of pavements (a) flexible, (b) semi-rigid and (c) rigid pavements. Modified from Plati [2]

When used as unbound aggregate, studies show that 100% RAP reduces the strength and increase the permanent potential deformation of the unbound bases [43]. Seferoğlu et al. [44] have found that the performance significantly increases with 3% PC treatment. According to the authors, RAP-3% PC and 20% RAP – 80% VA have similar deformation performance, which was 50% less than that of a conventional base course made with 100% VA.

The strain capacity appears to be also a function of the binder content present on the RAP particles [11]. Arshad and Ahmed [29] presented results from other authors who systematically agree that RAP addition to VA will (i) decrease the dry density, (ii) decrease the bearing capacity (measured by the California Bearing Ratio) and (iii) increase the resilient modulus

Roller compacted (RC), and lean concrete (LC) concrete with RAP aggregates are also an alternative for wearing course in semi-rigid pavements (see Figure 1). A 50% RAP replacement in RC concrete (fine and coarse aggregates) reduced the 28 days compressive strength and conversely increased the flexural strength [45]. According to Debbarma et al. [31], RAP aggregates also help reducing the permeable voids, water absorption in RC concrete, as well as lowering the cost per cubic meter in 46%. However, the cost reductions by employing RAP aggregates may be overtaken with the need to increase the (more costly) binder content. Singh [46] investigated dry lean concrete and observed that the mixes required at least 50% extra PC to compensate for the strength loss caused by 100% coarse RAP replacement.

The term full-depth reclamation (FDR) is found in the literature to designate stabilised bases composed of recycled RAP together with parts of underneath aggregates [9,47]. The study of Castañeda López et al. [48] shows that the additions of RAP in the cement-treated base as FDR may reduce the flexural strength but leads to more ductile behaviour. The fatigue life of cement-treated RAP depends much on the composition (PC and RAP content) and the thickness of the layer. Consequently, it is possible to use higher RAP content by playing with other variables (PC content and thickness).

2.3. RAP as aggregate in PC matrices

Most studies on RAP as an aggregate replacement choose between replacing either the fine or the coarse fraction (see Table 1). The level of replacement also varies, and a combination of RAP aggregates and natural aggregates is often employed. Overall, the employment of RAP aggregates in PC based matrices has a significant effect on both fresh and hardened state [49].

\[\text{resilient modulus} = \text{ratio of deviator stress to the resilient strain experienced by the material under repeated loading simulating traffic load} \]
<table>
<thead>
<tr>
<th>Author</th>
<th>Type of RAP</th>
<th>Replacement</th>
<th>w/c</th>
<th>Compressive Strength (MPa)</th>
<th>Splitting tensile strength (MPa)</th>
<th>Modulus (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Su et al. [35]</td>
<td>Coarse + Fine</td>
<td>0%</td>
<td>0.5</td>
<td>42.06</td>
<td>3.38</td>
<td>34.8 ELAS</td>
</tr>
<tr>
<td></td>
<td></td>
<td>20%</td>
<td>0.5</td>
<td>31.21</td>
<td>2.90</td>
<td>26.8 ELAS</td>
</tr>
<tr>
<td></td>
<td></td>
<td>40%</td>
<td>0.5</td>
<td>23.12</td>
<td>2.28</td>
<td>19.9 ELAS</td>
</tr>
<tr>
<td></td>
<td></td>
<td>70%</td>
<td>0.5</td>
<td>17.33</td>
<td>1.90</td>
<td>14.5 ELAS</td>
</tr>
<tr>
<td></td>
<td></td>
<td>100%</td>
<td>0.5</td>
<td>11.93</td>
<td>1.51</td>
<td>10.5 ELAS</td>
</tr>
<tr>
<td>Brand and Roesler [41]</td>
<td>Fine</td>
<td>100%</td>
<td>0.42</td>
<td>15.89</td>
<td>2.42</td>
<td>25.84 LONG</td>
</tr>
<tr>
<td>Shi et al. [50]</td>
<td>Coarse (different sources)</td>
<td>0%</td>
<td>0.40</td>
<td>33.61</td>
<td>4.39</td>
<td>25.98 TRAN</td>
</tr>
<tr>
<td>Thomas et al. [52]</td>
<td>Coarse</td>
<td>0%</td>
<td>0.47</td>
<td>73.8</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Abraham and Ransinchung [39]</td>
<td>Fine</td>
<td>0%</td>
<td>0.44</td>
<td>~51</td>
<td>4.9</td>
<td>-</td>
</tr>
<tr>
<td>Singh et al. [40]</td>
<td>Ref</td>
<td>0%</td>
<td>0.38</td>
<td>~43</td>
<td>4.6</td>
<td>-</td>
</tr>
<tr>
<td>Singh et al. [50]</td>
<td>Coarse/ Old</td>
<td>50%</td>
<td>0.38</td>
<td>35.3</td>
<td>2.6</td>
<td>-</td>
</tr>
<tr>
<td>Singh et al. [30]</td>
<td>Reference</td>
<td>100%</td>
<td>0.38</td>
<td>27.2</td>
<td>3.7</td>
<td>-</td>
</tr>
<tr>
<td>Debbarma et al. [31]</td>
<td>Reference</td>
<td>0%</td>
<td>0.37</td>
<td>~37</td>
<td>4.2</td>
<td>-</td>
</tr>
<tr>
<td>Huang et al. [51]</td>
<td>Reference</td>
<td>100%</td>
<td>0.5</td>
<td>37.7</td>
<td>3.21</td>
<td>-</td>
</tr>
<tr>
<td>El Euch Ben Said et al. [53]</td>
<td>Coarse + Fine</td>
<td>0%</td>
<td>0.6</td>
<td>30.1</td>
<td>3.1</td>
<td>31.1 ELAS</td>
</tr>
<tr>
<td></td>
<td></td>
<td>20%</td>
<td>0.6</td>
<td>23.8</td>
<td>2.8</td>
<td>28.9 ELAS</td>
</tr>
<tr>
<td></td>
<td></td>
<td>40%</td>
<td>0.6</td>
<td>20.7</td>
<td>2.3</td>
<td>24.6 ELAS</td>
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<td>16.7</td>
<td>2.2</td>
<td>22.3 ELAS</td>
</tr>
<tr>
<td></td>
<td></td>
<td>75%</td>
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<td>13.7</td>
<td>2.0</td>
<td>20.1 ELAS</td>
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<tr>
<td></td>
<td></td>
<td>100%</td>
<td>0.6</td>
<td>11.4</td>
<td>1.7</td>
<td>14.5 ELAS</td>
</tr>
<tr>
<td>El Euch Ben Said et al. [53]</td>
<td>Reference</td>
<td>20-100%</td>
<td>0.45</td>
<td>~34</td>
<td>28.52 ELAS</td>
<td></td>
</tr>
<tr>
<td>Papanastatious [54]</td>
<td>Fine</td>
<td>5-15%</td>
<td>0.45</td>
<td>~30-28</td>
<td>26.78-22.88 ELAS</td>
<td></td>
</tr>
<tr>
<td>Shatarat et al. [55]</td>
<td>Coarse</td>
<td>20-100%</td>
<td>0.57</td>
<td>~46</td>
<td>26.94-29.97 ELAS</td>
<td></td>
</tr>
<tr>
<td>Hossiney et al. [56]</td>
<td>Coarse + Fine</td>
<td>10-40%</td>
<td>0.53</td>
<td>38.58</td>
<td>3.73</td>
<td>32.95 ELAS</td>
</tr>
<tr>
<td></td>
<td></td>
<td>50%</td>
<td>0.37</td>
<td>40.0</td>
<td>6.3</td>
<td>44.6 ELAS</td>
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<tr>
<td></td>
<td></td>
<td>20%</td>
<td>0.37</td>
<td>37.2</td>
<td>4.7</td>
<td>37.4 ELAS</td>
</tr>
<tr>
<td></td>
<td></td>
<td>35%</td>
<td>0.37</td>
<td>32.7</td>
<td>3.4</td>
<td>31.9 ELAS</td>
</tr>
<tr>
<td>Brand et al. [37]</td>
<td>Coarse</td>
<td>0%</td>
<td>0.37</td>
<td>27.9</td>
<td>3.0</td>
<td>30.9 ELAS</td>
</tr>
</tbody>
</table>
Singh et al. [30] studied the incorporation of coarse and fine RAP into PC concrete and found a significant reduction in workability of samples. The strength reduction was more substantial with the employment of the fine fraction. When compared with concretes containing VA, the replacement of 100% coarse RAP reduced the slump by 29%, while 100% fine RAP reduced the slump by 100%. The authors attributed this to the absorption of the mixing water by the dust that accumulated on RAP during stockpiling. A reduction in workability was also observed by other authors [35,49], although opposite findings were also reported [37,50,51]. In general, it is still not clear how the following different parameters affect the workability of the fresh product: (i) hydrophobic properties and the high viscosity of the asphalt layer on the aggregate surface; (ii) high water demand of the adhered dust layer and (iii) the particle shape of the aggregates.

Through the production of RAP-PCC, it is possible to observe the formation of three interfacial transition zones (ITZ) in the microstructure: between (i) the aggregates and asphalt mortar, (ii) aggregates and cementitious mortar and (iii) asphalt mortar and cementitious mortar (Figure 2). The weakest ITZ is the one formed between asphalt mortar and cementitious mortar [30,53]. Brand and Roesler [36] observed that ITZ changed as the samples aged; overtime the samples with RAP developed higher porosity, larger ITZ width and fewer hydrates. The authors suggested that the high pH of the pore solution could leach organic compounds from the asphalt, which compromises the hydration process and formation of calcium hydroxide (CH) and calcium silicate hydrate (C-S-H). The consequence is the weakening of the ITZ over time.

Fracture samples with RAP have shown that most of the cracks propagated within the asphalt layer on the aggregate [41] instead of an adhesive failure of the asphalt-cement paste.

As a consequence, the use of fine and coarse RAP aggregates in the production of PC concrete can drastically reduce the mechanical properties, such as compressive strength, splitting tensile strength and elastic modulus, as well as increase the toughness of the material [30,35,39,40]. Concretes with 20-50% coarse aggregate replacement presented 30-60% compressive strength reduction [52].

Some authors observed that the used fine RAP to produce mortars could cause high shrinkage, reduction of 41.7% on compressive strength, 39.3% on flexural strength and 51.5% on splitting tensile strength [39,57]. Nevertheless, RAP-mortars also showed better ductility, toughness and crack resistance when compared to VA-mortars [57].

In general, the coarse RAP fraction is less detrimental to the mechanical properties [30]. However, the study of Debbarma et al. [31] on RC concrete, found that fine RAP mixes had a better compressive strength performance than the ones with coarse RAP. The improvement in strength was most likely due to the specific properties of the fine RAP used, such as low asphalt content and better gradation.

The inclusion of RAP on concretes and mortars reduces the splitting and flexural strength of the samples, which is more prominent when fine RAP is used [31,35,39]. However, the percentage reduction in flexural strength is lower than the compressive strength [37,50]. It is believed that better
Flexural performance is due to the visco-elastic nature of the asphalt coating, which also is responsible for an increase in the toughness of the mixes [51].

Shatarat et al. [55] tested concrete columns and observed a 31.06% reduction in the compressive load capacity in comparison with the control sample. The RAP samples also showed less cracking and lower spalling. The coefficient of thermal expansion and drying shrinkage does not seem to be impacted by the use of RAP aggregates [56].

2.3.1. Durability

The porosity of mortars increases in the presence of fine RAP aggregates due to the more porous and larger ITZ. Mercury intrusion porosimetry (MIP) analysis showed an increase in mesopores and macropores. According to Abraham and Ransinchung [58], the threshold diameters and large capillaries decreased up to 50% RAP replacement, followed by an increase of both after that, suggesting a higher risk for chemical ingress [58].

The presence of coarse RAP decreases the water absorption on concretes, the concentration of total permeable voids and sorptivity [30,31]. However, Abraham and Ransinchung [39,59] noted that this behaviour was in contradiction with the higher porosity of the RAP aggregates. The authors suggested that the decrease in sorptivity was due to the molten asphalt clogging the pores of the samples during oven drying. To avoid the melting of the asphalt present in RAP, the authors have adopted a different preparation procedure, and vacuum dried the samples at 35°C. Singh and Ransinchung [60] suggested oven drying at 48°C for eight days.

The inclusion of fine RAP can significantly impact the abrasion resistance of concrete samples; there is a 75% reduction on abrasion resistance when 100% RAP replacement is employed [61]. Still, samples produced with either coarse and fine RAP mixes produced enough abrasion resistance to be used as pavement's wearing course [31].

The replacement of VA with fine RAP had an impact on the pH of the hardened concrete; the pH reduces as the replacement level increase. This reduction is reported to be subtle (up to 3.8%), and all RAP-PC still present pH values above 12 [61]. No carbonation was observed after 120 days of moist curing [31,61].

Under sulphate attack, RAP-PC concrete samples also showed a higher mass and strength loss than the reference PC concrete, with rates increasing with RAP content [31,59]. Singh et al. [61] reported a weight loss of nearly 11.26% for concrete samples produced with 100% fine RAP replacement under H2SO4 exposure.

The freeze and thaw resistance of PC concrete is associated with the concrete’s ability to withstand the water expansion inside its pores and cracks during thermal cycling. It is also directly related to the material’s pore structure. In that sense, the higher percentage of air voids in RAP-PC concrete is beneficial and can help accommodate the expanding freezing water without damage to the matrix. Guo et al. [49] and Thomas et al. [52] reported an increase in freeze-thaw resistance when VA is replaced by RAP aggregates, while Brand et al. [37] observed a reduction.

2.4. Alkali activated materials as binders

The term alkali activation may be applied to the reaction between any aluminosilicate precursor and an alkali activator to produce a new binder [62–64]. Precursors are usually in powder form and may be occurring natural pozzolans, calcined clays or industrial by-product and wastes, such as granulated blast furnace slag, granulated phosphorus slag, steel slag, coal fly ash, volcanic glass, zeolite, metakaolin, silica fume and non-ferrous slags [62,65]. Depending on the Al and Ca content, AAM may be further classified as inorganic polymers or geopolymers (Figure 3), or simply as low-calcium (low-Ca) and high-calcium (high-Ca) AAM. As for activators, the most frequently used are sodium silicate, sodium hydroxide, sodium carbonate, potassium hydroxide and potassium silicate [66]. Hoy et al. [67,68] observed that the presence of sodium silicate in the alkali solution increased the speed of the alkaline activation. However, the ideal amount of activators may vary according to the type of precursor; and excess of activator results in loss of strength [69].

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2 Threshold diameter is the pore size through which mercury penetrates the bulk of the sample, i.e. the equivalent pore size corresponding to the steepest slope of the curve [112,113].
2.4.1. Ground Granulated Blast Furnace Slag (GGBFS) and Metakaolin (MK) based AAM

High-Ca AAM is mainly represented by the alkali-activation on GGBFS, while low-Ca AAM is commonly produced with pulverised fly ash (PFA) and metakaolin (MK). This section will compare the activation of GGBFS and MK as examples of high and low-Ca systems.

GGBFS is a latent hydraulic material and, as such, it requires water and a minimal amount of a catalyst or activator to show cementitious properties. When mixed with PC, GGBFS is activated by the calcium hydroxide and alkalis formed during the hydration of the cement. In the absence of PC, the activation is carried out using an alkali solution made with an alkali hydroxide, silicate, carbonate or sulphate, but combining sources (sodium hydroxide and sodium silicate) can yield even higher strength results [71]. Wang, Scrivener and Pratt [72], employed individual activators and concluded that sodium silicate solution with silica modulus\(^3\) from 1 to 1.5 achieved best mechanical performance. When using sodium hydroxide, the authors recommended keeping the Na\(_2\)O concentration within 3 and 5% by slag weight. They reported that lower Na\(_2\)O percentages might delay the activation process, while higher values may result in efflorescence and brittleness.

The chemical process from alkali-activation is entirely different from the reactions involved in PC hydration. Many researchers have contributed to a better understanding of the chemical mechanism, but the activation process is not yet fully understood. A full description of the reaction mechanism is beyond the scope of this study. Figure 4 outlines the key processes and products from alkali-activation.

The activation of any aluminosilicate starts with the dissolution of the solid particles by hydrolysis and produces aluminate and silicate species. The high pH of the medium accelerates the dissolution and quickly supersaturates the solution [73]. However, the activator’s pH plays a different role on calcium solubility (high-calcium systems). The high pH reduces the solubility of the calcium species as supersaturation is reached for portlandite [74]. This concentrated solution leads to the formation of a gel, and the systems continue to rearrange to form a three-dimensional aluminosilicate network [75]. During the nucleation step, ongoing reactions favour gel growth, and precipitation [65].

The main reaction product formed by alkaline activation of GGBFS is calcium (alumino)silicate hydrate (also known as C-A-S-H type gel), and secondary phases such as layered double hydroxides and an alkali aluminosilicate N-A-S-(H) type gel (can also show the substitution of potassium) [62,74,76,77]. The tetrahedrally coordinated silicate chains form the C-A-S-H gel are longer (compared to C-S-H gel from PC), and there is a substitution of the bridging silicon by aluminium. This substitution caused a charge imbalance that is compensated by the alkali ions in the gel [73].

\[^3\text{SiO}_2/\text{M}_2\text{O} \text{ molar ratio, where M is an alkali metal.}\]
According to the literature, an adequately designed alkali-activated slag shows improvements in many properties compared to PC – higher strength, more resistant to acids, sulphates or chlorides and fire resistance [65]. However, slag-based AAM may also present some issues such as rapid setting, high shrinkage, the formation of micro-cracks, the possibility of alkali-aggregate reaction and high efflorescence [71]. The addition of superplasticisers and water reducers designed for PC into AAM seems to have a negligible effect on workability and setting time [65].

Metakaolinite (Al₂Si₂O₇) also known as metakaolin (MK) is a product from the dihydroxylation of an aluminosilicate clay mineral kaolinite (Al₂(Si₂O₅)(OH)₄) at a temperature of between 450 - 800°C. During calcination, the kaolinite releases OH- from its structure, which becomes amorphous and reactive. MK has CaO as impurities; it is a system composed mainly of (Si+Al), while BFS is a (Si+Al+Ca) system [78]. Like other pozzolans, MK does not have cementitious properties, but when used with PC, it is a highly reactive pozzolan which reacts with the hydrated lime to form C-S-H [79]. The addition of MK to PC based systems can improve the mechanical strength, reduce the pore structure and mitigate some chemical attacks, such as alkali-silica reaction [80,81].

MK is also suitable for alkali activation yielding a material with high compressive strength, elastic modulus and lower porosity. Best properties are achieved in the presence of soluble silicates with the silica moduli of the final solution between 1 and 2 [74]. The reaction sequence of low calcium systems (green path on Figure 4) is similar to the one for high calcium, but the main reaction product is an alkaline aluminosilicate hydrate (N-A-S-H gel) with zeolite-like secondary phases. The N-A-S-H gel is considered as an amorphous three-dimensional framework of both SiO₄ and AlO₄ tetrahedrally coordinated and randomly interlinked by shared O atoms. The metal cations from the alkali solution (Na⁺ or K⁺) compensates the negative imbalance caused by the four-coordinated Al [82,83]. The absence of (or low content) calcium hinders the formation of crystalline phases associated with expansions (ettringite and gypsum), and the end products have better chemical stability [84]. When properly designed, the final geopolymeric product has a quicker setting time, higher compressive strength, better resistance to seawater and sulphate attack, and higher heat resistance than PC systems [64,84].

Nevertheless, the alkaline activation of MK represents a higher environmental impact than the activation of GGBFS. First, kaolin clay is a natural material, and its utilisation represents the depletion of natural resources. Secondly, the calcination of kaolinite demands the burning of combustible fuels, which then releases CO₂. MK activation mainly requires sodium silicate to provide the soluble silica to accelerate the hardening; however, the production of silicates is itself a heavy environmental burden [85]. Because of all that, GGBFS is the preferable aluminosilicate over MK, when the first is available. The mix of
GGBFS and MK (binary system) is a potential solution to achieve an end material with high compressive strength, lower porosity [86] and lower shrinkage than alkali-activated slag [87]. Binary systems show improvement in properties while still reducing the amount of alkaline activator needed for neat MK activation. Figure 5 shows that the activation of blended MK/GGBFS favours the formation of a denser matrix composed of C-A-S-H and N-A-S-H gels, which improves the overall properties.

![Figure 5. EDX spot analysis of MK+GGBFSS systems [86]](image)

2.5. RAP and alkali-activated materials

Many researchers have successfully studied the incorporation of RAP in base/subbase layers for pavement section; for high replacement levels, the loss on mechanical properties needs to be compensated by using stabilisers, i.e. agents that increase the compressive and impact strength, as well as the overall integrity of the layers (reduction of swelling, shrinkage, abrasion) [33,88]. It is possible to stabilise RAP using materials such as lime, cement, fly ash, blast furnace slag, rice husk ash, sugar cane bagasse ash, among others [30,69,89,90]. The stabiliser's content varies depending on the type of stabiliser employed, but it usually ranges from 4% to 30% in mass of RAP (see Table 2). While the high dosage of Ca-rich stabilisers is discouraged - it causes shrinkage, cracking and failure of the pavement - the use of low-Ca stabiliser may not impair sufficient strength. Therefore, sometimes, it is possible to employ alkali activation to meet the strength requirement for base materials [88]. The use of alkali activation in RAP base/subbase layers may help compensate the strength loss.

The literature on the alkaline activation of base/subbase pavement layers containing RAP materials is still limited. Hoy et al. [68] and Horpibulsuk et al. [91] presented results of alkali-activated base courses made with RAP and stabilised with high calcium (CaO >10%) PFA. The SEM and XRD analysis indicate that the main activation products were C-A-S-H and N-A-S-H gels and that the coexistence of both hydrates increased the strength, which was proportional to the Na_2SiO_3 content. The absence of silicates resulted in low geopolymerisation for RAP-PFA systems, as in conventional alkali-activated concretes [68]. The authors also found C-S-H as a hydration product in systems with a higher calcium content such as those with GGBFS [67].

These preliminary studies also conclude that similarly to the RAP-OPC systems, the presence of the asphalt layer is detrimental do the binding of RAP to the matrix. In other words, the RAP particles presented better bonding to the matrix for exposed aggregate surfaces (no asphalt layer). Hence, better performance may be achieved if a milling procedure guarantees a higher exposed aggregate area [32].

While studying the incorporation of RAP and PFA on different types of soil for road base and subbase layers, Adhikari [69] observed that an increase in the PFA and RAP content increased the strength of the geopolymer mixture up to 7 times compared with untreated soil. The elastic modulus and fracture energy also increased. However, when compared with OPC stabilised samples, the alkali-activated ones were more flexible and presented lower compressive strength and stiffness [92]. Other authors also observed the positive effect of increasing the PFA content [38,91]. Curing conditions also appear to have a significant impact on the mechanical properties, since it increases de dissociation (reactivity) of the phases. Alkali activated RAP samples cured for seven days at 40ºC presented approximatel.
## Table 2. Summary of the studies on alkali-activated matrices containing RAP (UCS: unconfined compressive strength)

<table>
<thead>
<tr>
<th>Author</th>
<th>RAP: VA</th>
<th>Stabiliser</th>
<th>Alkali activator</th>
<th>UCS 28 days (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jallu et al. [93]</td>
<td>60:40</td>
<td>PFA 20%</td>
<td>NaOH (3M)</td>
<td>0.50 - -6.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>NaOH:Na₂SiO₃ - 50:50</td>
<td>0.54 - -6.5</td>
</tr>
<tr>
<td>Hoy et al. [68,95]</td>
<td>100:0</td>
<td>PFA 20%</td>
<td>NaOH (10M)</td>
<td>- -3.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>NaOH:Na₂SiO₃ - 90:10</td>
<td>- -4.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>NaOH:Na₂SiO₃ - 60:40</td>
<td>- -4.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>NaOH:Na₂SiO₃ - 50:50</td>
<td>- -5.6</td>
</tr>
<tr>
<td>Saride; Avirneni; Challapalli [32]</td>
<td>80:20</td>
<td>PFA 20%-30%</td>
<td>NaOH</td>
<td>2-4% - -4.3 - 4.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>30%</td>
<td>NaOH</td>
<td>2-4% - -4.7 - 5.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>20%</td>
<td>NaOH</td>
<td>2-4% - -4.6 - 5.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>30%</td>
<td>NaOH</td>
<td>2-4% - -5.0 - 5.4</td>
</tr>
<tr>
<td>Mohammadinia et al. [92]</td>
<td>100:0</td>
<td>PFA 4%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.3-0.5</td>
</tr>
<tr>
<td></td>
<td>+BFS 2%-2%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.3</td>
<td>- 1.6</td>
</tr>
<tr>
<td></td>
<td>BFS 4%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.4</td>
<td>- 1.7</td>
</tr>
<tr>
<td>Mohammadinia et al. [38]</td>
<td>100:0</td>
<td>PFA 4%-16%</td>
<td>Na₂SiO₃/NaOH(8M)= 2.5</td>
<td>L/B =0.4</td>
</tr>
<tr>
<td></td>
<td>20%, 30%</td>
<td>NaOH (10M)</td>
<td>-</td>
<td>4 - 5</td>
</tr>
<tr>
<td></td>
<td>20%, 30%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.3</td>
<td>- 4 - 4.3</td>
</tr>
<tr>
<td></td>
<td>20%, 30%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.5</td>
<td>5 - 5.5</td>
</tr>
<tr>
<td></td>
<td>20%, 30%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.3-0.5</td>
<td>5.5 - 6</td>
</tr>
<tr>
<td>Horpibulsuk [91]</td>
<td>100%</td>
<td>PFA 10-20%</td>
<td>NaOH (8M)</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>20%, 30%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.3-0.5</td>
<td>4 - 4.3</td>
</tr>
<tr>
<td></td>
<td>10-20%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.3</td>
<td>1 - 3.0</td>
</tr>
<tr>
<td></td>
<td>10-20%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.3-0.5</td>
<td>1.5 - 2.2</td>
</tr>
<tr>
<td>Hoy et al. [67]</td>
<td>100:0</td>
<td>BFS 0%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0%</td>
<td>NaOH</td>
<td>1.6-5.38</td>
</tr>
<tr>
<td></td>
<td></td>
<td>5%-10%</td>
<td>NaOH (6-8M)</td>
<td>1.02-3.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0%</td>
<td>Na₂SiO₃/NaOH (0 -0.5)</td>
<td>1.62-4.98</td>
</tr>
<tr>
<td>Adhikari et al. [69]</td>
<td>0%</td>
<td>PFA 15%-25%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0%</td>
<td>NaOH:Na₂SiO₃</td>
<td>L/B =0.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0%</td>
<td>Na₂SiO₃/NaOH (0 -0.5)</td>
<td>1.02-3.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>15%-25%</td>
<td>NaOH (6-8M)</td>
<td>1.62-4.98</td>
</tr>
</tbody>
</table>
70% higher strength than reference samples cured at room temperature [38]. However, high-temperature curing is not practical for on-site applications. The flexural behaviour of the RAP-AAM bases can be improved with the use of geogrids by up to 2.54 times [93].

Wet-dry cycles are commonly used to simulate weather changes over time and estimate durability. Avirneni et al. [88] observed that activated PFA as stabilisers for RAP: VA mixes had an acceptable performance under aggressive wet/dry cycles, with less than 10% weight loss. Alkali activated systems presented lower weight loss on wet-dry cycling than non-activated ones. After six wet/dry cycles, the formation of macro- and micro-cracks caused a strength reduction, which was more significant for lower NaOH contents [94].

One great environmental concern when using wastes and alkali-activated materials is the possibility of contamination of soil and watercourses due to leaching. The durability of base layers may also be affected by leaching of stabilisers during its service life. Preliminary studies comparing the leaching of stabilised RAP with and without alkali activation have concluded that the environmental impact on soil and groundwater is minimal. The studies assessed the leaching of heavy metals, pH and sodium, concluding that the employment of an activator has reduced the leachability of metals from the aluminosilicate source [68, 88, 91].

3. Reflections from literature

Even with the current recycling efforts, the construction industry has been unable to consume all of the produced RAP, and more recycling alternatives are needed. Although great success is achieved when RAP is reused in asphalt layers [96–98], the same cannot be said for base and subbase layers. The use of RAP for unbound base layers represent a reduction in strength and attempts to compensate this reduction often means:

1. Blending it with virgin aggregate. Which reduces the number of recycled materials used and therefore the reduces the sustainability prospect of the solution.
2. Using binders or stabilisation. PC will add an environmental burden to the solution; supplementary cementitious materials (such as PFA, BFS and rice husk ash) take longer time to react and may not add sufficient strength.

The presence of the thin asphalt coating in the surface of the RAP aggregate is the main reason for the binder's poor performance. The asphalt weakens the ITZ, making it larger and more porous. RAP beneficiation methods targeting the increase of the exposed aggregate surface could represent a significant gain in strength. While the more porous ITZ could represent a gain in freeze and thaw resistance, it also compromises the durability by permitting the ingress of harmful substances. RAP beneficiation methods targeting the increase of the exposed aggregate surface could represent a significant gain in strength and durability of RAP-PCC.

The literature on RAP with AAM is very scarce, and some important factors remain unanswered. Similarly to RAP-PPC solution, the ITZ is also a reason for concern. However, it is unclear if different precursors and alkali solutions could improve de binding between the different ITZ sections. The use of a properly designed AAM may promote the recycling of RAP by increasing the early strength, creating a denser and more resistant (durability) matrix. The application of AAM is very similar to PC, and minor changes on the construction practice are expected. However, the costs associated with the transportation of the alkali solution must be considered.

3.1. Potential, challenges and research needs for future developments on RAP-AAM

This section covers important topics that have not been addressed so far for the future development of RAP incorporated alkali-activated materials. Nevertheless, they are essential to determine the potential application of those new materials by the pavement industry. The following section also highlights future research needs.

3.1.1. Cost and availability of local materials

Different from the employment of lime or Portland cement as binders for stabilisation of aggregates, AAM depends on the availability and costs of the alkaline solutions and precursors used (aluminosilicates). In that sense, the overall cost to develop RAP-AAM may surpass the benefits from some countries. Sodium silicate and sodium hydroxide, the most common activators used in AAM, are rather expensive chemicals in some countries, where AAM have barely become viable for most of the
possible applications [99]. Each country may have to select the available sources of aluminosilicate. Calcium-rich materials (industrial by-products or wastes) are preferred because they tend to reduce the demand for alkaline solutions. So, the employment of blast-furnace slag or high-calcium PFA may be an option to consider. Agricultural residues such as rice husk ash or sugarcane bagasse ash may be viable and inexpensive options for developing countries (Brazil, India, other Asian countries), as they are interesting raw materials for alkaline activation [100,101]. However, they are usually low calcium and low alumina reactive materials, which therefore depend on another source of $\text{Al}_2\text{O}_3$ and high amounts of activators.

The activation may be carried out using blended (or hybrid) systems, e.g. small amounts of lime or a reduced amount of OPC to promote the activation with a minimum amount of alkalis and enhance the final properties [102,103]. The study of those blended systems with RAP aggregates is not available, and it is a potential topic for investigation.

The activation of local clay materials as binders for RAP-AAM must also be considered and should be an object of future research. After all, clay minerals are the main constituent of soils, and the availability of this precursors should not represent an issue. The challenges of using natural clays as binders for AAM are related to their crystalline nature and consequently, low reactivity [104]. Soil stabilisation with alkaline materials has been studied, and many publications are now found in the literature [105–107].

The activation of soil clay minerals to bind RAP either in base or sub-base courses for flexible pavements or in semi-rigid (concrete) bases is a topic not explored as well.

Overall, it is paramount to assess the economic viability of AAM under two completely different perspectives:

- Stabilisation of RAP in base or subbase courses. In this case, the mechanical requirements are low and, therefore, low-reactively materials and small amounts of alkaline activators may be employed;
- RAP-alkali-activated concrete used as a rigid base for asphalt surface layers (semi-rigid solutions). This solution may demand higher mechanical performance and, hence, large quantities of activators in combination with highly-reactive precursors.

3.1.2. Performance and durability of RAP-AAM

The performance and durability of any RAP-AAM systems are closely associated with the amount and gradation of RAP used as aggregates, together with any beneficiation methods employed to the former. These variables determine the adhesion and ultimately, the mechanical performance of RAP to AAM matrices. The choice of low calcium or high-calcium precursors (and consequently the amount and type of activators) may also play an important role in the mechanical and durability properties. None of these topics has been systematically addressed so far in the published literature and represent essential research needs before the implementation of RAP-AAM pavement layers.

Long-term or accelerated durability tests employed to current pavement layers are also required to determine the durability of RAP-AAM under mechanical stresses (fatigue, rutting resistance) [13] as well as weathering conditions such as freeze-thaw cycling [94]. Needless to mention the importance of assessing the susceptibility to delamination of RAP-AAM concrete base from the asphalt surface layer in semi-rigid pavements, stabilising a comparison with traditional Portland cement concrete (PCC) [108].

Leaching of alkalis with consequent contamination of soil and groundwater are questions to be answered as well. Leaching is a function of the type of precursor and amount of alkalis used; in general, it should not present an issue if the alkali content is limited. On the other hand, alkali-activated matrices have proven to be suitable to encapsulate heavy metals and toxic waste [109]; thus, the leaching of undesirable species could even reduce, including some contaminants in the RAP aggregates.

3.1.3. Life cycle assessment (LCA) of RAP-AAM

Another object of future research is the life cycle assessment of RAP-AAM pavement layers. The environmental impact of AAM as a replacement for PC has been widely discussed [110,111], and the benefits are dependent on the precursor and amount of solution employed. The same applies to RAP-AAM, an LCA comparing RAP-AAM, traditional RAP-stabilised layers and Portland cement concrete base layers must be subject for future studies. LCA should be combined with the cost analysis to determine when RAP-AAM is viable. So far, it is not possible to say if the challenges to design and
specify RAP-AAM pavements would be both technically acceptable and economically feasible. Future research should definitively address these questions.

4. Conclusions

This paper presented the findings of a literature review on the use of RAP aggregates on cement bound systems. The investigation was an overview of the impact caused by using RAP aggregates on Portland cement and alkali-activated matrices. The main objective was to identify if RAP-AAM could be a suitable foundation layer of pavements while also proving more sustainable alternative (higher RAP replacement levels and lower environmental impact) than RAP-PC.

Similarly to RAP-PC, the biggest challenge facing the use of RAP-AAM is how to overcome the binding issues caused by the presence of asphalt. The asphalt coating on the aggregates reduces the mechanical properties and durability due to increased permeability. This may be addressed by physical or chemical pre-treatments of the RAP aggregates or by improvements on the binder.

The amount of research on RAP-AAM is limited; most studies used only PFA as a precursor, and still little is known about mechanical and durability properties. It is necessary to address how important factors, such as the effect of the type of aluminosilicate (low calcium or high calcium), type and amount of activator, neat or hybrid systems, as well and gradation, and amount and beneficiation of RAP could help improve the binding properties and the overall final material.

The employment of alkali-activated matrices to RAP aggregates depends on the availability and cost of precursors for AAM. Local materials may include industrial and agricultural residues and wastes and soils, which could be a source of clay materials for alkaline activation. The use of local materials is of great importance when aiming for sustainable practice in the long term.

AAM may be used to stabilise RAP particles, or they could be part of an AAM-RAP concrete base. The amount of binder and alkaline materials in the former is much lower. The choice of the system determines the mechanical properties, durability and environmental impact, which should be carried out via LCA and compared with current solutions.

The use of RAP-AAM is very promising for pavement sections due to their high early strength and increased durability of the alkaline binder. An optimised mix design could help compensate for the reduction in strength caused by the use of RAP granules. In addition, the low strength requirement of foundation layers could represent a lower need for alkalis and consequently lower environmental impact of this solution.

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