Title: Sewage Sludge Ash Characteristics and Potential for Use in Bricks, Tiles and Glass Ceramics

Author 1
Name and Qualifications: Ciarán J. Lynn BE, MSc
Affiliations: PhD doctoral researcher, University of Birmingham, UK
Address: School of Civil Engineering, University of Birmingham, Edgbaston, Birmingham B15 2TT
Email: CJL301@student.bham.ac.uk

Author 2 (Corresponding Author)
Name and Qualifications: Prof. Ravindra K. Dhir OBE, BSc, PhD, CEng, MIMMM, HonFICT, HonFICI, FGS
Affiliations: Professor, University of Birmingham, UK
Address: School of Civil Engineering, University of Birmingham, Edgbaston, Birmingham B15 2TT
Email: r.k.dhir@bham.ac.uk

Author 3
Name and Qualifications: Dr. Gurmel S. Ghataora BEng, PhD, MIMMM, MILT, MMGS, MIGS
Affiliations: Senior lecturer, University of Birmingham, UK
Address: School of Civil Engineering, University of Birmingham, Edgbaston, Birmingham B15 2TT
Email: G.S.GHATAORA@bham.ac.uk

ABSTRACT
The characteristics of sewage sludge ash (SSA) and its use in ceramic applications pertaining to bricks, tiles and glass ceramics have been assessed using the globally published literature in the English medium. It is shown that SSA possess similar chemical characteristics to established ceramic materials and under heat treatment achieves the targeted densification, strength increases and absorption reductions. In brick and tile applications, technical requirements relating to strength, absorption and durability are achievable, with merely manageable performance reductions with SSA as a partial clay replacement. Fluxing properties of SSA facilitate lower firing temperatures during ceramics production, though reductions in mix plasticity leads to higher forming water requirements. SSA glass ceramics attained strengths in excess of natural materials such as granite and marble and displayed strong durability properties. The thermal treatment and nature of ceramic products also effectively restricted heavy metal leaching to low levels. Case studies, predominantly in bricks applications, reinforces confidence in the material with suitable technical performances achieved in practical conditions.

Key Words: Bricks, Glass Ceramics, Sewage Sludge Ash, Tiles
INTRODUCTION

Sewage sludge is a by-product of wastewater treatment. With ever reducing limits on the allowable biodegradable landfilled fraction [European Community (1999)], combined with a ban on its disposal at sea [European Community (1991)] and restrictions on its spreading on agricultural land, management of the waste has shifted towards more environmentally friendly treatment methods such as recycling and incineration.

The incineration process leads to a 90% reduction in volume, leaving behind residual sewage sludge ash (SSA), whilst the high temperatures involved in the combustion process reduce the toxic components remaining in the ash.

The ever increasing global emphasis on sustainability and with it, developing the greater use of sustainable materials, calls for the use of all the so-called waste materials as valuable resources and in this case, establishing SSA as a viable secondary construction material can contribute greatly to realising this target. Indeed, the use of SSA in ceramic products, such as bricks and tiles and glass ceramics offers great promise given the vast size of the industry, whilst the thermal processes involved in production may also have beneficial impacts on diminishing concerns regarding leaching of heavy metals in SSA.

Much research has been undertaken worldwide, studying the characteristics of SSA and its use in ceramic applications, though in its current form carried out by different authors, in different countries and with different approaches, the research remains largely fragmented and not utilised to the fullest. As such, a critical evaluation of the collective data, including technical and environmental assessment, along with the work from case studies, can be useful and timely, both to advance the safe and sustainable use of SSA and to lessen future repetitive work and squandering of resources.

THE PROJECT

A critical analysis, evaluation and repackaging of the global literature published in the English medium, is undertaken, studying the physical and chemical characteristics of SSA and its potential use in ceramics applications, pertaining to construction products such as bricks, tiles and glass ceramics.

A comprehensive search using SSA related keywords in over thirty search engines and databases yielded a total of 196 publications, produced since 1972 from 33 countries across Europe (92 publications), Asia (80), North America (14), Africa (4), South America (4) and Australia (1), with key contributions coming from Taiwan (31 publications), Japan (26), UK (24), Spain (15), German (14) and USA (13).

Due to the large volume of literature, publications providing data solely relevant to the characteristics of SSA were not cited, to avoid overwhelming the messages delivered in the text. However, the lists of these publications are provided as supplementary data.

SEWAGE SLUDGE ASH CHARACTERISTICS

Analysis of the characteristics of SSA is based on data extracted from 168 publications from 30 countries, produced since 1972, with the yearly publication breakdown presented in Figure 1. The level of research carried out on SSA is on the rise, with the majority of work undertaken in the last 10 years, reflecting the growing importance of incineration and recycling of secondary materials.
Physical Characteristics

SSA has been reported as a free-flowing, odourless material with a yellowish-brownish colour. Analysis of the fineness of SSA, along with particle size distribution curves presented in Figure 2, show that the material is composed predominantly of fine sand and silt size particles, suggesting natural suitability as filler material in ceramics, or as a replacement of clay after grinding. The grinding process can also help to regulate the particle size spread evident in Figure 2, given the precise dimensional tolerances and configuration requirements set for ceramic products such as clay masonry units.

Specific gravity values ranging from 1.8-2.9 have been reported, though the data is skewed towards the upper end of this range due to one particularly low value [Jamshidi et al. (2012)], with average and standard deviation values of 2.6 and 0.3 respectively (45 SSA samples).
Density has been shown to increase with higher incineration temperatures, whilst the percentage of the denser heavy metal components contained in the ash is another key determining factor in the overall density.

The low ratio of bulk specific gravity (average of 0.9 for 13 samples) to particle specific gravity is indicative of a porous material. This is consistent with observations of the morphology of SSA that revealed irregularly shaped particles with rough surface textures and a porous microstructure. This porosity promotes high water absorption properties and indeed an average absorption value of 18% has been determined from the literature (8 samples), though it should be noted that two extreme values of 1.8% and 48% have been reported by Al-Sharif and Attom (2014) and Environmental & Water Technology Centre of Innovation (2012), respectively.

**Chemical Characteristics**

A ternary plot of the main oxides in SSA samples from the literature is presented in **Figure 3**, along with the composition of other established ceramic materials given as references. It is evident that SSA contains high contents of SiO₂ and Al₂O₃, oxides that are commonly present in ceramic materials, though the material has a higher CaO content, with an average value of 14.5%.

Fe₂O₃ is another main oxide found in SSA (average content of 11.6%) that is also commonly found in ceramic materials. This component, along with other oxides such as CaO and P₂O₅, can lead to added benefits due to fluxing properties, which can lower the melting temperature of the mixture during sintering and reduce energy costs. Besides the above similarities, the contents of P₂O₅ and SO₃, with average values of 13.7% and 3.5% determined respectively for SSA, are much greater than what is typically present in standard materials used in ceramics production.

![Figure 3: Analysis of the main oxides of SSA](image-url)
Low amounts of organic matter have remained in SSA after incineration. An average LOI (loss on ignition) of 3.9% has been calculated based on the sourced literature (49 samples) and though irregularly high values of approximately 15% have been reported in some cases [USEPA (1972)], the organic content of SSA is generally lower than materials such as clay and shale. The additional thermal treatment involved in ceramics production should also minimize the presence of organics in the final product.

Quartz, calcite and hematite have been identified as the most abundant minerals present in SSA, whilst glass contents ranged from 35-75%. Many other iron oxides, iron phosphates, calcium phosphates, aluminosilicates and aluminium phosphates have been reported to a lesser extent, depending on factors such as the raw sludge composition, additives used during processing or incineration and the incineration temperature. SSA possesses some similarities to brickwork feedstocks and clays, which typically contain minerals such as quartz, calcite, hematite, kaolinite, feldspar and mica. The interaction and mineralogical evolution of SSA with these materials under firing will have a key impact on the properties of the resultant products.

An analysis of the element composition, according to the reported data is presented in Table 1, including both toxic and non-toxic components. The variability evident from the coefficient of variation results is quite striking and can be attributed for the most part to differences in wastewater treatments systems and the incineration conditions around the world.

If current EU landfill leaching classification limits [European Community (2003)] were applied, SSA would not meet the classification as an inert material due to the leached contents of heavy metals such as Cd, Cr, Cu, Ni, Pb and Zn. However, the high temperatures involved in the firing process and make-up of the resultant ceramic products can significantly downgrade the risks associated with leaching of harmful components from SSA, while at the same time the material is serving as a valuable construction material and also reducing landfilling demands, thus making this particular use very appealing.

The presence of active soluble salts in raw materials, in general, is a recognised concern in ceramics production that can negatively affect the manufacturing process, product aesthetics and durability. In this regard, maximum limits on the Na⁺, K⁺ and Mg²⁺ soluble salt contents are set in EN 771-1 for the produced clay masonry units. Though not frequently covered in the literature, two publications have reported on the content of soluble salts in SSA as varying from 0.15 - 1.52%. The effects of these components in SSA on the resultant ceramic products is considered in the next Section on ceramics applications. However, if required, chemical additives are available to restrict the movements of these components [Anderson and Skerratt (2003)], though the potential variability in the soluble salts contents and the associated variability in the chemical additives required would have to be considered.
Table 1: Element composition of SSA

<table>
<thead>
<tr>
<th>ELEMENT</th>
<th>SAMPLE NUMBERS</th>
<th>MEAN mg/kg</th>
<th>S.D. mg/kg</th>
<th>CV %</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>TOXIC</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fe</td>
<td>33</td>
<td>75664</td>
<td>54231</td>
<td>72</td>
</tr>
<tr>
<td>Al</td>
<td>31</td>
<td>45771</td>
<td>27026</td>
<td>59</td>
</tr>
<tr>
<td>Zn</td>
<td>88</td>
<td>2988</td>
<td>3506</td>
<td>117</td>
</tr>
<tr>
<td>Cu</td>
<td>89</td>
<td>1826</td>
<td>3078</td>
<td>169</td>
</tr>
<tr>
<td>Ba</td>
<td>11</td>
<td>1909</td>
<td>724</td>
<td>38</td>
</tr>
<tr>
<td>Cr</td>
<td>77</td>
<td>585</td>
<td>1069</td>
<td>183</td>
</tr>
<tr>
<td>Sr</td>
<td>8</td>
<td>399</td>
<td>150</td>
<td>38</td>
</tr>
<tr>
<td>Pb</td>
<td>86</td>
<td>344</td>
<td>445</td>
<td>129</td>
</tr>
<tr>
<td>Ni</td>
<td>69</td>
<td>228</td>
<td>371</td>
<td>163</td>
</tr>
<tr>
<td>V</td>
<td>15</td>
<td>147</td>
<td>181</td>
<td>123</td>
</tr>
<tr>
<td>Co</td>
<td>15</td>
<td>150</td>
<td>180</td>
<td>119</td>
</tr>
<tr>
<td>Se</td>
<td>9</td>
<td>8</td>
<td>12</td>
<td>162</td>
</tr>
<tr>
<td>Sb</td>
<td>10</td>
<td>32</td>
<td>25</td>
<td>77</td>
</tr>
<tr>
<td>As</td>
<td>36</td>
<td>26</td>
<td>43</td>
<td>164</td>
</tr>
<tr>
<td>Cd</td>
<td>70</td>
<td>18</td>
<td>61</td>
<td>343</td>
</tr>
<tr>
<td>Hg</td>
<td>37</td>
<td>2</td>
<td>3</td>
<td>148</td>
</tr>
<tr>
<td><strong>NON-TOXIC</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Si</td>
<td>13</td>
<td>108667</td>
<td>54575</td>
<td>50</td>
</tr>
<tr>
<td>P</td>
<td>31</td>
<td>63084</td>
<td>37316</td>
<td>59</td>
</tr>
<tr>
<td>Ca</td>
<td>26</td>
<td>67176</td>
<td>35165</td>
<td>52</td>
</tr>
<tr>
<td>Na</td>
<td>20</td>
<td>11055</td>
<td>20125</td>
<td>182</td>
</tr>
<tr>
<td>Mg</td>
<td>22</td>
<td>13038</td>
<td>6053</td>
<td>46</td>
</tr>
<tr>
<td>K</td>
<td>19</td>
<td>8663</td>
<td>6327</td>
<td>73</td>
</tr>
<tr>
<td>Ti</td>
<td>11</td>
<td>2454</td>
<td>3196</td>
<td>130</td>
</tr>
<tr>
<td>Mn</td>
<td>30</td>
<td>1149</td>
<td>753</td>
<td>66</td>
</tr>
<tr>
<td>Zr</td>
<td>10</td>
<td>377</td>
<td>251</td>
<td>67</td>
</tr>
<tr>
<td>Sn</td>
<td>12</td>
<td>128</td>
<td>153</td>
<td>119</td>
</tr>
<tr>
<td>Ag</td>
<td>10</td>
<td>216</td>
<td>198</td>
<td>92</td>
</tr>
<tr>
<td>Cl</td>
<td>28</td>
<td>1526</td>
<td>3149</td>
<td>206</td>
</tr>
<tr>
<td>Mo</td>
<td>32</td>
<td>29</td>
<td>31</td>
<td>107</td>
</tr>
</tbody>
</table>

USE IN CERAMIC APPLICATIONS

Characterisation as a Ceramic Material

Although much work has been done on the use of SSA in specific applications such as bricks, tiles and glass ceramics, preliminary information on the generic response of the material to the ceramics production process is limited [Anderson and Skerratt (2003), Cheeseman et al. (2003), Lin et al. (2006b), Merino et al. (2005), Merino et al. (2007) and Okufuji (1990)].

As a ceramic material, a basic understanding of the behaviour of SSA during the heating/cooling process is required. In addition, properties such as dimensional stability, density, strength, thermal properties, water absorption, and durability are of importance. From an initial thermal analysis, softening and melting point temperatures for SSA have
ranged from 1210-1280°C and 1250-1300°C respectively [Merino et al. (2005) and Okufuji (1990)]. Apparent density results are presented in Figure 4 (a) for SSA specimens produced at various maximum heating temperatures and in addition the effects of heating dwell times, compaction pressures and preparation treatments that have varied in these studies, are also portrayed.

![Graph showing apparent density and maximum heating temperature and compressive strength after thermal treatment.](image)

Figure 4: Relationship between (a) apparent density and maximum heating temperature and (b) apparent density and compressive strength after thermal treatment.

**Figure 4 (a)** shows that with densification of the material during heating, involving shrinkage of pore spaces, identified by the SEM analysis of Lin et al. (2006b), SSA attains a maximum average density of about 2.25 kg/m$^3$ at 1000-1200°C with coefficient of variation under 5%. It appears that variations in heating dwell time, compaction pressure and preparation methods had little effect on the peak density. As previously stated in the characteristics section, variation in the temperature required for the SSA to attain maximum density is known to be influenced by the fluxing properties of oxides such Fe$_2$O$_3$, CaO and P$_2$O$_5$. Beyond the optimum firing temperature (i.e. point of peak density), a bloating phenomenon is instigated leading to the creation of large isolated pores and therefore a steep decline in density, as is evident in **Figure 4 (a)**.

Strength increases are related to density by a power relationship as shown by the trendline in **Figure 4 (b)**. The correlation seems to remain consistent despite variations in firing temperature and time, compaction pressure and preparation methods. As the maximum densities achieved for the range of test setups were all relatively similar, a reasonable assessment of the maximum strengths attainable with SSA can be gained from **Figure 4 (a)** and (b) combined.
Control of the water absorption properties is important with SSA, given the porous and absorptive nature of unprocessed material and can be particularly crucial in certain situations such as with externally exposed ceramic units. Encouragingly, the water absorption properties have been shown to be inversely related to density and compressive strength and reduce drastically from 30% to less than 1% [Cheeseman et al. (2003), Lin et al. (2006b) and Merino et al. (2007)], as density increases to a maximum. Absorption rises at temperatures past optimum firing conditions due to the bloating effect.

There appears to be a lack of research on the durability of SSA as a ceramic material per se, though its performance in this regard is catered to with the specific products such as bricks, tiles and glass ceramics.

In ceramics production, a level of consistency in the raw materials is a clear requirement, particularly given the importance of chemical composition on the optimum firing temperature (Figure 4 (a)). For fired SSA samples taken over a six week period from the same incineration plant, tensile strengths results ranged from 6-15 MPa [Anderson and Skerratt (2003)], though the variability has been attributed to differences in the particle size distributions and not chemical compositions, as all samples demonstrated comparable fusion/melting behaviour. This is helpful as the particle size distribution of SSA can be regulated with sieving. In addition, when SSA is used in small proportions, these inconsistencies would likely not significantly affect the overall performance.

Of additional interest, the inclusion of various clays and glass alongside SSA [Merino et al. (2007)], provides an idea of the behaviour expected with SSA as a partial component in bricks, tiles or glass. Montmorillonite and illite clays led to improvements in density and compressive strength, though this was not the case with kaolin clay due to its refractory properties. The fluxing properties of powdered flat glass lowered the temperature required for densification to occur and as such, can be useful for reducing energy requirements.

**Bricks**

Bricks offer great promise as an outlet for SSA use, with approximately 1300 billion units produced worldwide per annum [Clean Air Task Force (2010)]. The available experimental data on the effects of SSA on the brick production [Anderson (1999), Anderson (2002), Lin and Weng (2001), Luo and Lin (2003), Tay (1987) and Trauner (1993)] shows that:

- SSA lowers the plasticity of the mix, though the associated decrease in the bonding ability can be overcome using additional tempering water and the effects on extrusion behaviour are not significant at low SSA contents.

- The fluxing properties of calcium, iron, phosphorus and potassium in SSA benefit the fusion behaviour, thus facilitating lower firing temperatures.

**Table 2** is presented with the aim of evaluating the performance of SSA bricks specifically relative to the standard UK and USA specifications, covering BS EN 771-1 and Publicly Available Specification PAS 70 (UK) and ASTM C62-13a and ASTM C216-15 (building and facing bricks in the USA).

**Appearance:** No adverse effects on the aesthetics of the bricks have been reported, with one exception [Anderson (2002)], where “drier scum” was visible due to the soluble salts in SSA.
This is a recognised problem in general with ceramic raw materials and can be overcome using chemical additives to fix the salts.

Table 2: Overview of the UK and USA bricks performance specifications

<table>
<thead>
<tr>
<th>UK SPECIFICATIONS</th>
<th>USA SPECIFICATIONS</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Appearance</strong></td>
<td></td>
</tr>
<tr>
<td>PAS-70 - guide on appearance including colour consistency, cracks and damage</td>
<td>ASTM C216-15 - limits on chips, cracks, imperfections, broken bricks, colour, efflorescence</td>
</tr>
<tr>
<td><strong>Compressive strength</strong></td>
<td></td>
</tr>
<tr>
<td>BS EN 771-1 - No minimum strength. Category I - &lt; 5% of units below declared strength, otherwise Category II. UK National Annex Engineering bricks - Class A &gt; 125 N/mm², Class B &gt; 75 N/mm²</td>
<td>ASTM C216-15/C62-13a - Min strength: Average - 20.7 (SW), 17.2 (MW) and 10.3 (NW) MPa, Individual units - 17.2 (SW), 15.2 (MW) and 8.6 (NW) MPa.</td>
</tr>
<tr>
<td><strong>Freeze thaw</strong></td>
<td></td>
</tr>
<tr>
<td>BS EN 771-1 - Declare resistance category: F0 - passive exposure, F1 - moderate exposure, F2 - severe exposure</td>
<td>ASTM C216-15/C62-13a - Freeze thaw test - SW - no unit has weight loss &gt; 0.5% and no unit develops a crack of length in excess of the units least dimension</td>
</tr>
<tr>
<td><strong>Soluble salts</strong></td>
<td></td>
</tr>
<tr>
<td>BS EN 771-1 - Categories: S0 (completed protected) no limits. S1 (normal exposure) Na + K &lt; 0.17%, Mg &lt; 0.08%. S2 (prolonged saturation) Na + K &lt; 0.06%, Mg &lt; 0.03%</td>
<td>No requirements</td>
</tr>
<tr>
<td><strong>Water absorption</strong></td>
<td></td>
</tr>
<tr>
<td>BS EN 771-1 - Requires water absorption to be declared. In UK National Annex - Engineering brick class A/DPC1 &lt; 4.5%, class B/DPC2 &lt; 7.0% (5hrs boiling test.)</td>
<td>ASTM C216-15/C62-13a - Limits (5hrs boiling) - Average 17% (SW), 22% (MW), no limit (NW). Individual - 20% (SW), 25% (MW), no limit (NW).</td>
</tr>
</tbody>
</table>

Compressive Strength: SSA bricks can be produced to comply with ASTM C216-15/C62-13a and BS EN 771-1 for strength for specific grades, suggesting that the reductions with SSA contents can be manageable (Figure 5) and that the sensitivity of strength development to firing temperature, as evident from the results of Trauner (1993), is of greater influence than SSA use.

Figure 5: Brick compressive strength with SSA replacement level
Freeze Thaw: The results of Trauner (1993) show that the ASTM C216-15/C62-13a limit for severe weathering class bricks of 0.5% weight loss in freeze-thaw testing can be achieved with bricks manufactured using up to 30% SSA. The weight loss behaviour appears to mimic that of water absorption.

Soluble Salts: Although discussed in terms of the impacts on aesthetics, the soluble salts contents in SSA bricks and their potential effects on brick durability have not been measured.

Water Absorption: Whilst the effect of SSA, shown in Figure 6, appears to be somewhat inconsistent, variability in absorption for the control bricks, from 0.03 - 14%, again suggests a greater dependence on pore structure development during firing rather than SSA use as can be seen from the results of Trauner (1993) for firing temperatures of 1040°C and 1120°C. In relation to the performance set out in the Standards, the 5 hour boiling water test data [Lin and Weng (2001) and Tay (1987)] suggest that SSA bricks can satisfy the limits (< 17% for severe weathering) for building and facing bricks (ASTM C216-15/C62-13a). For the more stringent engineering class and damp proof course (DPC) bricks (< 7% absorption), SSA is perhaps more suited for use at low contents.

Figure 6: Brick water absorption performance with SSA replacement

Although drying shrinkage is not referred to in the UK or USA Standards, the data show that SSA benefitted the quality of the bricks in this regard, due to its lower swellability and plasticity compared to clay.

From an economic standpoint, though the value of brick making materials is quite low, with landfilling costs rising, the prospects for SSA use in bricks can only improve. It is suggested that the costs can be further reduced by situating the incineration plants and brick making facilities closer. The fluxing properties of SSA may also lead to energy savings in the production process, though additional tempering water and anti-scum agents may be required. As a sign of the interest in SSA, a number of additional publications without available experimental data, have discussed SSA use, such as: Donatello and Cheeseman (2013), Johnson et al. (2014), Kadir and Mohajerani (2011), Smol et al. (2015), Tay and Show
(1992), Vouk et al. (2015) and Wienerberger (2012). Case studies in this area are also dealt with in Section 6.

**Tiles**

SSA use in tiles, both glazed and unglazed, manufactured using pressuring procedures and focused as a clay replacement, has been predominantly examined in the Asian countries and evaluated in line with Chinese Standards [CNS (1999)], with no major production problems arising due to SSA [Chen and Lin (2009), Dayalan and Beulah (2014), Kaneko et al. (1992), Lin et al. (2005a), Lin et al. (2007b), Lin et al. (2008) and Okufuji (1990)]. Whilst unglazed tiles at times exceeded the 16% water absorption limit of CNS (1999), Figure 7(a), taking clay tiles as a zero baseline (Figure 7 (b)) with the exception of Lin et al. (2005a), showed that SSA had only minor effect in this respect, particularly at low SSA contents. Glazing has been found to reduce absorption, at a rate that was similar for both control and SSA tiles.

![Figure 7: (a) Tile water absorption performance with SSA replacement and (b) zeroed tile water absorption displaying the isolated effects of SSA](image)

Despite bending strength reductions, at an average rate of 0.5% per 1% SSA, all products with up to 40% SSA satisfied wall and floor tile earthenware requirements for this of 5.9 and 9.8 MPa [CNS (1999)] respectively [Chen and Lin (2009), Lin et al. (2005a), Lin et al. (2007b) and Lin et al. (2008)]. Nano-SiO$_2$ additions (tested up to 3%) as expected improved strength, but at greater rates with SSA, particularly at higher contents (30-50% SSA), though became ineffective at temperatures exceeding 1150°C. Glazing has been shown to further improve strength with SSA use (Lin et al, 2008), though this remains to be confirmed (Lin et al. (2005a).

All tiles (up to 50% SSA) fulfilled the CNS (1999) abrasion limit of 0.1g, despite drops in resistance with SSA [Chen and Lin (2009), Lin et al. (2005a), Lin et al. (2007b) and Lin et al. (2008)]. Glazing greatly improved abrasion (optimal glaze contents of 0.03-0.1%) and more effectively for SSA tiles compared to controls. Nano-SiO$_2$ additives enhanced abrasion properties at a comparable rate at all SSA contents, though only effectively at temperatures from 1050-1100°C.
SSA had no significant effect on acid-alkali or ageing resistance tests, although 0.1 \( \text{g/cm}^2 \) glaze has been needed for both control and SSA tiles for categorisation as “good” in both aspects [Lin et al. (2005a) and Lin et al. (2008)]. Due to fluxing properties, red glaze colorant (Fe\(_2\)O\(_3\)), has been most beneficial to durability, whilst purple glaze (MnO\(_2\)) has been least effective.

SSA did not significantly affect firing shrinkage of tiles. In addition, weight loss on ignition (WLOI) reduced with SSA, as the organic matter had already been burnt off during sewage sludge incineration.

**Glass Ceramics**

SSA use in glass ceramics requires it to be part of the process of combining melting with controlled crystallization, achieving the sealing, low porosity and fabrication advantages of glass, together with the strength and resistance properties of ceramics [Donatello and Cheeseman (2013), Endo et al. (1997), Hnat et al. (1999), Isa (2011), Park et al. (2003), Rawlings et al. (2006), Smol et al. (2015), Suzuki et al. (1997), Wystalska et al. (2013), Yoon and Yun (2011), Zhang et al. (2013) and Zhang et al. (2015)]. Table 3, which presents the mix details, shows that CaO has been commonly used as a conditioning additive to achieve anorthite and diopside crystalline phases known for high strength. Silica sand, soda ash and dolomite have also been incorporated at times, whilst in one case [Yoon and Yun (2011)] SSA was mixed directly with waste glass without melting.

**Table 3: Mix details, heat treatment and performances of SSA glass ceramics**

<table>
<thead>
<tr>
<th>REFERENCE</th>
<th>MIX</th>
<th>HEATING</th>
<th>CS, MPA</th>
<th>BS, MPa</th>
<th>HARDNESS</th>
<th>TEC, 10(^{-6})/°C</th>
<th>DENSITY, g/cm(^3)</th>
<th>WA, %</th>
<th>AR, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Endo et al. (1997)</td>
<td>SSA+CaO</td>
<td>Tm 1490°C, Tc 1000-1100°C</td>
<td>164</td>
<td>n/a</td>
<td>n/a</td>
<td>67</td>
<td>n/a</td>
<td>0</td>
<td>0.1</td>
</tr>
<tr>
<td>Park et al. (2003)</td>
<td>SSA+CaO</td>
<td>Tm 1500°C, Tc 1050°C; 2h, 1150°C, 3h</td>
<td>n/a</td>
<td>75-92</td>
<td>5860-6230 MPa (V)</td>
<td>74-83</td>
<td>2.87-2.93</td>
<td>0</td>
<td>&lt; 2</td>
</tr>
<tr>
<td>Suzuki et al. (1997)</td>
<td>SSA+CaO+Dol+SiO(_2)+SSA</td>
<td>Tm 1400°C, Tc 1000-1200°C; 1-8h</td>
<td>n/a</td>
<td>n/a</td>
<td>5-6 (M)</td>
<td>67</td>
<td>3</td>
<td>n/a</td>
<td>0.1</td>
</tr>
<tr>
<td>Yoon and Yun (2011)</td>
<td>SSA+waste glass</td>
<td>No melting, Tc 850-1050°C, 1h</td>
<td>210-270</td>
<td>119-156</td>
<td>4790-5560 MPa (V)</td>
<td>n/a</td>
<td>2.32-2.69</td>
<td>n/a</td>
<td>0.13-0.14</td>
</tr>
<tr>
<td>Wystalska et al. (2013)</td>
<td>SSA, SSA+Dol</td>
<td>Tm 7 T, 871, 986, 1150°C, 1h</td>
<td>n/a</td>
<td>n/a</td>
<td>6.5 (M)</td>
<td>n/a</td>
<td>n/a</td>
<td>n/a</td>
<td>n/a</td>
</tr>
<tr>
<td>Zhang et al. (2013)</td>
<td>SSA+CaO</td>
<td>Tm 1500°C, Tc 977°C; 1-3h</td>
<td>n/a</td>
<td>n/a</td>
<td>n/a</td>
<td>2.19-2.30</td>
<td>1.55-2.30</td>
<td>n/a</td>
<td>n/a</td>
</tr>
<tr>
<td>Zhang et al. (2015)</td>
<td>SSA+SiO(_2)+CaO</td>
<td>Tm 1500°C, Tc 870, 945, 1065°C for 1-3h</td>
<td>167-247</td>
<td>77-118</td>
<td>n/a</td>
<td>2.37-2.88</td>
<td>0.42-1.02</td>
<td>n/a</td>
<td>n/a</td>
</tr>
</tbody>
</table>

**NATURAL MATERIALS**

<p>| | | | | | | | | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Marble</td>
<td></td>
<td>118</td>
<td>6-22</td>
<td>3-4</td>
<td>80</td>
<td>2.6-2.8</td>
<td>0.2</td>
<td>2.5</td>
<td>10.3</td>
</tr>
<tr>
<td>Granite</td>
<td></td>
<td>130</td>
<td>6-30</td>
<td>5-6 (M), 5500 MPa (V)</td>
<td>2.7</td>
<td>0.35</td>
<td>1</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Tm = melting temperature, Tc = crystallization temperature, Dol = dolomite, SA = soda ash, CS = compressive strength, BS = bending strength, TEC = thermal expansion coefficient, WA = water absorption, AR = acid resistance, V = Vickers hardness, M = Mohs hardness.

Glass melts have been produced at approximately 1500°C, though this dropped to 1400°C in the study undertaken by Suzuki et al. (1997) using four other constituents alongside SSA. During subsequent heating, nucleation occurred at 760-800°C (optimal duration of 1 hour), with iron sulphide colloids forming as nucleating agents. Optimal crystallization occurred at
temperatures from 1000-1150°C, forming anorthite, diopside and at times wollastonite crystals, resulting in high strengths.

The resultant glass ceramic properties, also presented in Table 3, show that SSA products achieved compression and bending strengths well in excess of typical marble and granite values. Hardness values were within the range expected for glass (5-7 Mohs) and were comparable or out-performed the reference natural materials. Under optimal thermal conditions, densities similar to marble and granite have been achieved. The products showed no excessive susceptibility to thermal expansion or acid attack, whilst low water absorption values have also been reported.

The colour of SSA glass ceramics ranged from dark graphite to brown depending on crystallization phases and with polishing, a beautiful high quality marble-like look has been achieved [Suzuki et al (1997)].

ENVIRONMENTAL ASSESSMENT

Though the data available on brick kiln emissions is quite limited, the fact that SSA comes from an already regulated incineration treatment somewhat eases this environmental concern. In a brick manufacturing trial incorporating 5% SSA, emissions monitored over 2 days did not exceed limits specified by the Environmental Protection Act (1990) [Anderson et al. (2002)].

Heavy metal leaching results for SSA itself and in ceramic products, presented in Table 4, have been determined from toxicity characteristics leaching procedure (TCLP) tests, though this method is intended to determine if a waste is hazardous, based on the EPA regulatory limits. With no leaching limits given for ceramic materials, Japanese environmental quality standards for soil [Ministry of the Environment (1991)] and EU drinking water limits [European Community (1998)] have also been included in Table 4 as indicators of performance.

Table 4 shows that:

- Leaching from SSA itself was below the hazardous classification limits, though exceeded the soil and drinking water limits in most cases.

- Sintering led to some reductions in the leaching, though cadmium concentrations remained above the soil and drinking water limits.

- Of added interest, acid neutralisation tests on sintered SSA, revealed increased susceptibility to leaching under strongly acidic conditions (pH of 3) [Cheeseman et al. (2003)].

- The leaching of tiles and glass ceramics with SSA reduced dramatically compared to the untreated material and with the cautionary exception of nickel, fell below the drinking water limits.

Thus, it would appear that the heat treatment involved and nature of ceramic products appears to make the safe use of SSA potentially viable.
Table 4: Leachability of SSA ceramic products (Note: Underline - exceeds EU drinking water parameters, bold - exceeds Japanese soil quality regulations)

<table>
<thead>
<tr>
<th>TEST PRODUCTS AND REFERENCES</th>
<th>HEAVY METAL LEACHABILITY, mg/L</th>
</tr>
</thead>
<tbody>
<tr>
<td>SSA</td>
<td>Cd</td>
</tr>
<tr>
<td>Lin et al. (2005a)</td>
<td>0.03</td>
</tr>
<tr>
<td>Lin et al. (2006b)</td>
<td>0.03</td>
</tr>
<tr>
<td>Zhang et al. (2015)</td>
<td>-</td>
</tr>
</tbody>
</table>

**Sintered SSA**
Lin et al. (2006b)
- 600°C, 2hrs: 0.03, <0.014, 0.4, - <0.016, 5.49
- 800°C, 2hrs: 0.02, <0.014, 0.4, - <0.016, 4.55
- 1000°C, 2hrs: 0.02, <0.014, 0.57, - <0.016, 2.82

**Tiles**
Lin et al. (2005a)
- 45% SSA + 55% clay, 1050°C: 0.002, 0.039, 0.991, 0.123, - 0.196

**Glass Ceramics**
Zhang et al. (2015)
- SSA + CaO/SiO₂ additives, 945°C, - 0.01 0.11 0.06 0.01 1.14
- SSA + CaO/SiO₂ additives, 1065°C, - 0.02 0.08 0.05 0.01 1.02
Endo et al. (1997)*
- SSA + CaO additive, 1100°C (A) <0.01 0.002 - - <0.01 -
- SSA + CaO additive, 1100°C (B) <0.01 <0.05 - - <0.01 -

**Limits**
EPA hazardous regulatory limits 1 5 15 15 5 -
Japanese Soil Quality Regulations 0.01 0.05 - - 0.01 -
EU Drinking Water Parameters 0.005 0.05 2 0.02 0.01 -

*Leaching method not specified.

**CASE STUDIES**
Case studies on SSA in ceramics have focused mainly on brick applications and the details of work done and the findings described in Table 5. The material has been used in quite a variety of ways in these projects, including as a filler component in place of sand together with clay, or as the sole raw material in brickmaking.

Table 5 indicates that effective practical use of SSA in ceramics has been achieved, with the products attaining satisfactory strength, absorption and durability properties and indeed, as a filler component, the material appears to be an upgrade over sand. Issues such as scumming, have been overcome by limiting the water absorption levels [Okuno and Takahasi (1997)], whilst increased forming water requirements have been counteracted by combining SSA with another waste material (water treatment residue) that on its own had a problematically high water content [Anderson et al. (2002)].
### Table 5: Description of work undertaken and findings on ceramics case studies with SSA

<table>
<thead>
<tr>
<th>Reference</th>
<th>Work Undertaken</th>
<th>Findings</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anderson et al. (2002)</td>
<td>UK. 200000 experimental bricks with SSA, WTR and CY from 0-10%, with clay.</td>
<td>After experimentation with mix designs, a final mix of 5% of each of SSA, WTR and CY achieved satisfactory production speed, mechanical, durability and aesthetic properties.</td>
</tr>
<tr>
<td>Anderson et al. (1996)</td>
<td>UK. Trial bricks with 8% SSA replacing sand based on a commercially used feedstock.</td>
<td>SSA is an upgrade over sand, with bricks achieving greater mechanical performance, no adverse effects on colour. Increased tempering water and staining were flagged as future possible issues.</td>
</tr>
<tr>
<td>DEFRA (2007)</td>
<td>UK. General overview of the use of wastes in bricks in the UK.</td>
<td>States that SSA is in use in 1 plant in UK at the time, though no figures provided. Fluxing agent, filler and colourant are listed as possible functions of SSA in brickmaking.</td>
</tr>
<tr>
<td>Mödinger (2002)</td>
<td>Italy. Industrial production was commencing with ceramic matrix composites using SSA.</td>
<td>Results not provided, yet it is reported that the reduced plasticity associated with SSA, would limit its use to lower contents.</td>
</tr>
<tr>
<td>Okuno et al. (2004)</td>
<td>Japan. Environmental and economic LCA of the previous project with 100% SSA bricks</td>
<td>Brickmaking LCA: Environmental – 0.076 kg CO$_2}$/ kg dewatered sludge used. Energy cost - 4.27¥ / kg dewatered sludge. Energy costs calculated exceeded the product sellable prices (2.24¥/kg dewatered sludge).</td>
</tr>
<tr>
<td>Petavratzi (2007)</td>
<td>UK. Pilot scale brick production with 2.5 and 5% SSA.</td>
<td>SSA demonstrated suitability as a filler or sand replacement. It was indicated that greater amounts of tempering water would be required with SSA. Scumming was present, though at an acceptable level.</td>
</tr>
<tr>
<td>Petavratzi and Barton (2007b)</td>
<td>UK. Industry study on brick manufacture, including the use of waste materials.</td>
<td>Similarly to the DEFRA report, states that SSA is in use in 1 plant in the UK, though further details are not provided.</td>
</tr>
<tr>
<td>Smith (2014)</td>
<td>UK. Reviews the use of non-primary clay materials in UK brickmaking</td>
<td>564 and 470 t of SSA used in brickmaking in 2005 and 2006 respectively, 0 usage from 2007-2010. Outlines many positives of using secondary materials including reduced energy usage and carbon footprint.</td>
</tr>
</tbody>
</table>

WTR – water treatment residue, CY – carpet yarn

Reservations about the environmental and economic competitiveness have been raised based on high CO$_2$ production and past energy costs in excess of the market value of bricks made from 100% SSA [Okuno et al. (2004)]. However, these economic conditions are subject to change, driven by the increasing importance of sustainability and continuous improvement of the thermal technologies to be more suited to these secondary materials. In addition, perhaps the gentle incorporation of SSA at low contents can be a more economically viable option rather than as the entire feedstock.

The performance-based approach now adopted in the EU standards also offers greater opportunity for the use of materials such as SSA, rather than past material-based specifications. Other important benefits that can encourage SSA use such as reduced landfill and extraction costs and conserving natural resources, are well known.
CONCLUSIONS

Analysis of the data suggests that SSA use in ceramics pertaining to bricks, tiles and glass ceramics is feasible, with potential for wide application. The specific findings are given as follows:

i. The fineness of SSA suggests natural suitability as filler material. Its irregularly shaped rough textured particles and porous microstructure leads to high absorption properties. SSA shares the same main oxides (SiO$_2$, Al$_2$O$_3$ and CaO) and minerals (quartz and calcite), that are commonly found in ceramic materials, whilst the fluxing properties of CaO, Fe$_2$O$_3$ and P$_2$O$_5$ can benefit fusion during heating. Heavy metals in SSA give rise to leaching concerns that must be controlled, whilst soluble salts present can potentially affect product aesthetics. SSA achieves optimal densification and associated high strengths and low absorption properties during firing at 1000-1200°C.

ii. In brick production, the fluxing properties of SSA facilitates lower firing temperatures, though additional forming water has been needed due to decreased plasticity. Strength and absorption requirements for building and facing bricks, have been satisfied with up to 50% SSA. Dried scum has been visible on occasion, but can be managed with anti-scum agents. Bricks with up to 30% SSA satisfied the most severe freeze-thaw resistance limits.

iii. No production problems arose with up to 50% SSA in both unglazed and glazed tiles. Despite drops in strength, products with up to 40% SSA satisfied wall and floor tile earthenware requirements, whilst effects on absorption appeared to be minor. Abrasion resistance requirements were satisfied for all SSA tiles, whilst acid-alkali and ageing resistance and firing shrinkage properties were not significantly affected. Heavy metal leaching is controlled effectively in tiles, falling predominantly below EU drinking water limits.

iv. Melting at 1500°C, nucleation at 760-800°C and crystallization at 1000-1150°C has resulted in optimal glass ceramics performances with SSA. Strengths exceeded typical values for granite and marble. SSA products displayed suitable durability and exhibited a high quality look when polished. Heavy metal leaching concentrations have also been effectively reduced to low levels in glass ceramics.

v. Case studies, predominantly on bricks, demonstrated satisfactory technical performances with SSA in practical situations, though reservations have been raised about the economic viability in a specific case with SSA used as the entire brick feedstock. Issues such as scumming have been overcome by limiting absorption levels, whilst SSA has been combined with a high water content material (water treatment residue) to counteract higher forming water requirements.

REFERENCES


Anderson M, Elliott M and Hickson C. 2002 Factory-scale proving trials using combined mixtures of three by-product wastes (including incinerated sewage sludge ash) in clay building bricks. *Journal of Chemical Technology and Biotechnology*, 77, 345-351


Chen L and Lin D F. 2009 Applications of sewage sludge ash and nano-SiO₂ to manufacture tile as construction material. *Construction and Building Materials*, 23, 3312-3320.


USEPA. 1972 Sewage sludge incineration, Report no. EPA-R2-72-040, PB 211 323.


**SUPPLEMENTARY DATA**

**Appendix A: Material Description**


**Appendix B: Fineness and Particle Size Distribution**


Donatello S, Tong D and Cheeseman C R. 2010 Production of technical grade phosphoric acid from incinerator sewage sludge ash (ISSA), Waste Management, 30, 1634-1642.


Sato Y, Oyamada T and Hanahara S. 2013 Applicability of sewage sludge ash (SSA) for paving materials: A study on using SSA as filler for asphalt mixture and base course material. Third International Conference on Sustainable Construction Materials and Technologies, August 2013, Kyoto, Japan.


Appendix C: Density


Sato Y, Oyamada T and Hanehara S. 2013 Applicability of sewage sludge ash (SSA) for paving materials: A study on using SSA as filler for asphalt mixture and base course material. Third International Conference on Sustainable Construction Materials and Technologies, August 2013, Kyoto, Japan.


Appendix D: Morphology


Appendix E: Absorption


Appendix F: Oxide Composition


Latosinska J. 2014 The evaluation of the impact of sewage sludge ash modification on leaching of heavy metals. *Advances in Civil and Environmental Engineering, 1*(1), 27-42.


Morais L C, Dweck J, Campos V and Buchler P M. 2009 Characterization of sewage sludge ashes to be used as a ceramic raw material. Chemical Engineering Transactions, 17, 1813-1818.


Stark K, Plaza E and Hultman B. 2006 Phosphorus release from ash, dried sludge and sludge residue from supercritical water oxidation by acid or base. Chemosphere, 62, 827-832.


Appendix G: Loss On Ignition


**Appendix H: Mineralogy**


Kjersgaard D. 2007 The reuse of bio ash for the production of concrete. A Danish case study. IWA Specialist Conference on Wastewater Biosolids, 24-27 June, Moncton, New Brunswick, Canada.


Zhang Z, Zhang L, Yin Y, Liang X and Li A. 2015 The recycling of incinerated sewage sludge ash as a raw material for CaO-Al$_2$O$_3$-SiO$_2$-P$_2$O$_5$ glass ceramic production. *Environmental Technology*, 36(9), 1098-1103.

**Appendix I: Trace Elements**


Ebbers B, Ottosen L M and Jensen P E. 2015 Comparison of two different electrodialytic cells for separation of phosphorus and heavy metals from sewage sludge ash. *Chemosphere*, 125, 122-129.


Latosinska J. 2014 The evaluation of the impact of sewage sludge ash modification on leaching of heavy metals. Advances in Civil and Environmental Engineering, 1(1), 27-42.


Morais L C, Dweck J, Campos V and Buchler P M. 2009 Characterization of sewage sludge ashes to be used as a ceramic raw material. Chemical Engineering Transactions, 17, 1813-1818.


Schaum C, Cornel P and Jardin N. 2011 Phosphorus recovery from sewage sludge ash – a wet chemical approach, Technische Universität Darmstadt, Germany. Available from


Tay J H and Show K Y. 1997 Resource recovery of sludge as a building and construction material – A future trend in sludge management. Water Science and Technology, 36(11), 259-266.


Vogel C and Adam C. 2011 Heavy metal removal from sewage sludge ash by thermochemical treatment with gaseous hydrochloric acid. Environmental Science & Technology, 45, 7445-7450.


Zhang Z, Zhang L, Yin Y, Liang X and Li A. 2015 The recycling of incinerated sewage sludge ash as a raw material for CaO-Al$_2$O$_3$-SiO$_2$-P$_2$O$_5$ glass ceramic production. Environmental Technology, 36(9), 1098-1103.