Properties of lightweight aggregate produced by rapid sintering of incinerator bottom ash

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Abstract

The fraction of municipal solid waste incinerator bottom ash with a particle size less than 8mm has been milled, formed into pellets and rapidly sintered in a rotary furnace at temperatures between 900 and 1080 °C. The effect of sintering temperature on density, water absorption and crushing strength has been determined. Sintering at temperatures between 1000 and 1050 °C produces pellets with physical properties comparable to Lytag, a commercially available lightweight aggregate manufactured from sintered pulverised fuel ash. Major crystalline phases present in milled bottom ash were quartz (SiO2) and calcite (CaCO3), while sintered pellets contained diopside (CaMgSi2O6), wollastonite (CaSiO3) and clinoenstatite (Mg2Si2O6). Leaching of heavy metals from sintered bottom ash pellets in water and under acid conditions (leachate pH 2–7) has been investigated. Rapid sintering at relatively low temperatures significantly reduced leaching in water compared to milled ash. Pb and Zn are leached under aggressive acid conditions (leachate pH 3) with 30–40% of the total present available for leaching. The results indicate that relatively simple processing of the finer fraction of incinerator bottom ash allows this problematic waste to be manufactured into lightweight aggregate with potential uses in a range of construction products and geotechnical applications. Current economic drivers for this in the UK are discussed.

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1. Introduction

The role of waste incineration is expected to increase in the UK and many other countries where the population density is high and availability of landfill is limited. Although incineration in modern energy from waste (EfW) plants reduces the volume of municipal solid waste (MSW) by up to 90%, this still produces significant quantities of incinerator bottom ash (IBA). The UK Environment Agency recently reviewed the fate of IBA produced by EfW plants in England and Wales. The 11 plants operating between 1996 and 2000 processed 8% of approximately 27.6 million tonnes of MSW generated each year, and this resulted in 642,088 tonnes of IBA in 2000. Seventy nine percent of this was sent direct to landfill, while the remainder was processed prior to use as bulk fill in applications such as embankments or as a substitute normal weight aggregate in asphalt (UK Environment Agency Report, 2002).

Solid waste management in industrialised countries increasingly aims to reduce the amount of waste requiring landfill, by developing viable reuse applications so that wastes are beneficially used as resources (Chang et al., 1999; Woolley et al., 2001). IBA is a heterogeneous mix of ceramic materials such as brick, stone, glass, ferrous and non-ferrous metals and other non-combustible inorganic and residual organic matter (Chimenos et al., 1999; Wiles, 1996; Zevenbergen et al., 1994). Given the possible increasing reliance on waste incineration, developing new, higher value reuse applications for this material is an important research area. In particular, as cities such as London and SE England continue to develop there is an enormous requirement for construction materials. Beneficial reuse of wastes in construction replaces materials that would otherwise need to be extracted from the environment and this has associated sustainability benefits. In addition, key drivers such as the increasing costs of waste disposal and extraction of natural aggregates are making processing wastes such as IBA into new construction products potentially viable.

2. Lightweight aggregate production from IBA

The production of lightweight aggregate (LWA) represents a particularly attractive reuse application for IBA (Owens and Newman, 1999a, 1999b). Most natural aggregates have particle densities of 2.4–2.8 g cm$^{-3}$, typically 2.6 g cm$^{-3}$, while LWA have particle densities of 0.8–2.0 g cm$^{-3}$. As a result, LWA are used for the production of lightweight concrete, lightweight blocks and other lightweight construction products and have additional benefits associated with low density such as high insulation and high thermal inertia. They are also increasingly used in a number of other applications including lightweight geotechnical fill, insulation products, soil engineering, hydro-culture, drainage, roof gardens and filters.

There are many different lightweight aggregates currently available. These can be either naturally occurring low-density materials such as pumice, scoria, volcanic cinders or diatomite or they can be manufactured by thermally treating expanding clays, shale, siliceous rock or slate, such as Liapor, Optiroc, Buildex, Stalite, Haydite, Perlite, Norlite and Solite. There are also LWA manufactured from industrial by-products such as fly ash and paper mill sludge, fly ash, sewage sludge and clay, and expanded blast furnace slag. Lytag is the...
leading LWA produced in the UK and this is manufactured from sintered fly ash (pulverised fuel ash) the by-product from coal fired power stations (Owens, 1993).

Ideally individual lightweight aggregate pellets should have:

- a strong but low density, porous, sintered ceramic core;
- a dense continuous surface layer to inhibit ingress of water that should be pozzolanic to produce a strong aggregate-cement bond in concrete;
- a near-spherical shape to improve fresh concrete properties.

The aim of this research was to produce a technically viable LWA by investigating the physical properties and leaching of rapidly sintered pellets made from milled IBA. Leaching studies are important to assess potential adverse environmental impacts associated with reuse of IBA.

The manufacture of LWA from IBA is potentially viable in the UK because:

(i) the cost of disposing of IBA to landfill is increasing due to the Landfill Tax and the requirements of the EU Landfill Directive;
(ii) the costs of primary (natural) aggregates have increased due to the imposition of an Aggregates Levy that is charged on each tonne of extracted aggregate;
(iii) there is a shortage of readily available aggregate in some regions of the UK and particularly where extensive house building is planned such as London and SE England. Natural aggregates are increasingly being transported over large distances to this region with implications for sustainability;
(iv) EfW plants producing IBA tend to be situated in urban areas close to where extensive construction activity is occurring. A potential source of LWA is therefore potentially available close to where these materials are most needed;
(v) the development of low-cost waste derived LWA for use in construction offers opportunities for developing products with improved thermal and sound insulation properties.

Sintering studies have been reported on the fly ash generated by incineration of MSW (Mangialardi, 2001; Wang et al., 2001, 2002). This is a hazardous material and there has been much less work reported on sintering IBA as this is generally much less problematic (Bethanis et al., 2002a; Cheeseman et al., 2003). It has been mixed with glass cullet and a range of other industrial wastes and used as a raw material for the production of glass–ceramics (Barbieri et al., 2000a, 2000b; Ferraris et al., 2001; Romero et al., 2001). These processes involve high temperature melting followed by subsequent heat treatment to control the microstructure of the resulting materials rather than sintering. The processing used in this work is different and produces different types of materials. LWA has previously been produced from IBA using an innovative rotary kiln (Wainwright and Cresswell, 2001), although limited materials characterisation data was reported.

3. Materials and methods

3.1. Incinerator bottom ash

IBA was collected from an ash recycling plant associated with a major EfW facility situated in SE England. This uses conventional mass-burn incineration technology, processes
approximately 420,000 tonnes of waste annually and generates 35 MW of electricity. The IBA is transported off-site to the ash recycling plant, where it is aged prior to sorting into different size fractions for potential reuse. The material with a particle size less than 8 mm is problematic, as there are currently no commercially viable reuse applications available for this material, although it represents approximately 45% by weight of the total IBA. Samples of this fraction were collected and dried prior to use in these experiments.

3.2. Sample milling and characterisation

Sub-samples of the <8 mm IBA were milled to reduce the particle size distribution using a tungsten carbide Tema mill (Gy–Ro, Glen Creston Ltd.). This is a rapid and aggressive milling technique in which the ash is effectively crushed between rapidly vibrating rings. The resulting fine powder was thoroughly mixed to ensure homogeneity and the particle size distribution determined by laser diffraction in the range between 0.4 and 900 μm (Beckman Coulter LS 100). The composition of milled IBA was analysed in triplicate using inductively coupled plasma atomic emission spectroscopy (ICP-AES, Perkin Elmer 3580B) following digestion of 5 g homogenised samples using lithium metaborate and tetraborate flux fusion (Ingamells, 1970).

3.3. Preparation of LWA pellets

A controlled amount of water (~24%) was added to the milled IBA powder to give a mix consistency that allowed formation of approximately spherical, 8–10 mm diameter pellets. These were dried overnight at 105 °C and fired at various temperatures by passing along the length of a rotary electric tube furnace (Carbolite). The rotating tube had an internal diameter of 7.7 cm and was 150 cm long with a 90 cm heated zone. The speed of rotation was fixed at 2.8 revolutions/min and the angle of tilt was kept constant at 2°. In order to control pellet traverse through the furnace, a series of baffles were fitted into the tube. The average pellet traverse time under these conditions was 10 min 35 s (standard deviation, 2 min, n = 30).

3.4. Physical property testing

The dry density (apparent specific gravity) and water absorption of sintered milled IBA pellets were determined by measuring the dry mass \( m_{\text{dry}} \), immersed mass \( m_{\text{imm}} \) and 24 h saturated surface-dry mass \( m_{\text{sat}} \) of between 10 and 20 pellets sintered at each temperature. Using Archimedes’ principle, individual particle densities were calculated from (Neville, 1994):

\[
\frac{m_{\text{dry}}}{m_{\text{sat}} - m_{\text{imm}}} \times 2 \, \text{cm}^{-3}
\]

and the 24 h water absorption value calculated from:

\[
\frac{m_{\text{sat}} - m_{\text{dry}}}{m_{\text{dry}}} \times 100
\]
Individual pellets have been loaded to fracture between two parallel plates as shown schematically in Fig. 1. Stress analysis has shown that when a sphere is tested in this way on two diametrically opposed points the compressive strength $S$ of the specimen is given by (Li et al., 2000; Yashima et al., 1987):

$$S = \frac{2.8P_c}{\pi X^2}$$

where $X$ is the distance between loading points, that is the sphere diameter, and $P_c$ is the fracture load. Mean values of the compressive strength were calculated from tests completed on at least 10 pellets prepared at each sintering temperature.

The physical properties (particle density ($n = 12$), water absorption ($n = 12$) and compressive strength ($n = 60$)) of commercially manufactured Lytag aggregate pellets have also been determined using identical test techniques to those for sintered IBA pellets.

3.5 Mineralogy and microstructural analysis

Milled and sintered IBA samples were analysed by X-ray diffraction (XRD, Philips PW1830 diffractometer fitted with PW1820 goniometer) on samples ground to less than 150 μm, using Cu Kα radiation, at an accelerating voltage of 40 kV and current of 40 mA.

Fracture surfaces of selected sintered IBA pellets were gold coated and examined using scanning electron microscopy (SEM, JEOL JSM-T220A).

3.6 Leach testing

The pH dependent leaching of heavy metals was investigated for milled IBA and IBA pellets sintered at 1050 °C using the acid neutralisation capacity (ANC) test combined with leachate analysis by ICP-AES (Stegemann and Côté, 1991). The sintered IBA pellet samples
were initially ground to less than 150 µm prior to leach testing. A series of 5 g sub-samples were mixed with 30 ml solutions of varying acidity, ranging from distilled water to 2.0N HNO₃ over 11 equal increments. Samples and leachants were mixed for 48 h in sealed containers on a rotary extractor, before being centrifuged. The leachate was then extracted by filtering through a 0.45 µm membrane filter and the pH measured before being acidified with 10% volume HNO₃, prior to metal analysis.

4. Results

4.1. Particle size distribution of milled IBA

The effect of milling the <8 mm IBA for 1, 2 and 4 min on the particle size distribution is shown in Fig. 2. The $d_{50}$ value for the as-received ash was 136.9 µm, and this did not change significantly during the first minute due to the nature of the milling process. However, $d_{50}$ values after 2 and 4 min milling were 21.0 and 12.6 µm, respectively, and the 2 min milled ash was selected for use in subsequent IBA pellet sintering experiments.

4.2. Chemical composition of milled IBA

The elemental composition of the milled IBA is included in Table 1. Major cationic elements, apart from Si present at high concentrations (>10 000 mg/kg) included Ca, Al, Na and Fe. Heavy metals of environmental concern present at relatively high concentrations (>100 mg/kg) included Cu, Zn, Pb and Cr.
Table 1
Average composition of milled bottom ash, and leaching (mg kg\(^{-1}\) DS) in distilled water (0 meq/g) from milled <8 mm IBA and IBA pellets rapidly sintered at 1050 °C

<table>
<thead>
<tr>
<th></th>
<th>Total (mg kg(^{-1}) DS (s.d.))</th>
<th>Leached (mg kg(^{-1}) DS)</th>
<th>Leached (%)</th>
<th>Leached (mg kg(^{-1}) DS)</th>
<th>Leached (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Leached pH = 10.2</td>
<td>Leached pH = 9.7</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Na</td>
<td>19307 (91)</td>
<td>2721</td>
<td>14.1</td>
<td>&lt;d.l.</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>K</td>
<td>3545 (48)</td>
<td>1299</td>
<td>36.6</td>
<td>0.01</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Mg</td>
<td>&lt;d.l.</td>
<td>&lt;0.01</td>
<td>0.3</td>
<td>&lt;d.l.</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Ca</td>
<td>5033 (503)</td>
<td>2134</td>
<td>8.8</td>
<td>1.1</td>
<td>0.1</td>
</tr>
<tr>
<td>Al</td>
<td>40261 (563)</td>
<td>483</td>
<td>1.2</td>
<td>11.1</td>
<td>0.3</td>
</tr>
<tr>
<td>Fe</td>
<td>72167 (602)</td>
<td>&lt;d.l.</td>
<td>&lt;0.01</td>
<td>&lt;d.l.</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Zn</td>
<td>3288 (42)</td>
<td>&lt;d.l.</td>
<td>&lt;0.01</td>
<td>11.4</td>
<td>0.4</td>
</tr>
<tr>
<td>Cu</td>
<td>5930 (43)</td>
<td>34.3</td>
<td>0.6</td>
<td>0.8</td>
<td>0.1</td>
</tr>
<tr>
<td>Pb</td>
<td>802 (64)</td>
<td>&lt;d.l.</td>
<td>&lt;0.01</td>
<td>&lt;d.l.</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Cr</td>
<td>217 (15)</td>
<td>&lt;d.l.</td>
<td>&lt;0.01</td>
<td>2.4</td>
<td>1.1</td>
</tr>
<tr>
<td>Cd</td>
<td>40 (3)</td>
<td>&lt;d.l.</td>
<td>&lt;0.01</td>
<td>0.3</td>
<td>0.7</td>
</tr>
</tbody>
</table>

* <d.l. = below detection limit of ICP-AES analysis.

4.3. Physical properties of sintered pellets

The effect of firing temperature on the density of sintered IBA pellets is shown in Fig. 3a. Pellets sintered below 1000 °C were weak and ‘chalky’. Sintering between 1000 and 1050 °C produced well-sintered pellets with consistent average densities, typically in the range between 1.4 and 1.6 g cm\(^{-3}\). Pellets sintered at 1060 °C showed some surface cracking and sintering above 1080 °C caused pellets to agglomerate and stick in the tube furnace. The results indicate that rapid sintering over a fairly broad temperature range (50 °C) can produce IBA pellets with densities comparable to, although slightly higher than those of Lytag.

Fig. 3b shows the effect of sintering temperature on water absorption of IBA pellets. The water absorption reduced with increasing temperature, indicating a reduction in open, water accessible porosity. The results were comparable to, and at higher sintering temperatures lower than, those of Lytag.

Fig. 3c shows the effect of sintering temperature on the compressive strengths of sintered IBA. Average strengths of IBA pellets sintered at all temperatures were lower than those obtained for Lytag. However, given that Lytag is a particularly strong LWA, the strengths obtained for sintered IBA would not preclude the use of the material as LWA.

4.4. Microstructural characterisation of milled and sintered IBA

XRD data for milled IBA is given in Fig. 4 (bottom trace). This indicates that quartz (SiO\(_2\)) and calcite (CaCO\(_3\)) are the major crystalline phases present together with some hematite (Fe\(_2\)O\(_3\)) and gehlenite (Ca\(_2\)Al\(_2\)SiO\(_6\)). This is consistent with previously reported
Fig. 3. Effect of firing temperature on the properties of sintered IBA pellets, (a) density (apparent specific gravity); (b) 24 hour water absorption and (c) compressive strength determined from testing individual pellets. Solid lines are mean data. Dashed lines are plus and minus one standard deviation.
Fig. 4. X-ray diffraction data for milled IBA and IBA sintered at 1080 °C.

Fig. 5 shows SEM micrographs of IBA sintered at 1020 and 1050 °C. Sintering at 1020 °C results in material containing significant glassy phase with isolated, irregularly pores. Samples sintered at 1050 °C appeared to contain approximately spherical pores, typically 5–10 μm in diameter. These are believed to form when the residual glass viscosity is low enough for gas-forming decomposition reactions to produce the voids observed.

data on the mineralogy of IBA (Bethanis et al., 2004; Dykstra et al., 1999; Kirby and Rimsidt, 1993).

XRD data for IBA sintered at 1030 °C is given in Fig. 4 (top trace). This material contained wollastonite (CaSiO3), diopside (CaMgSi2O6) and clinoenstatite (Mg2Si2O6) as the principal mineral phases, together with reduced levels of quartz and some hematite. A number of the XRD peaks detected could not be unambiguously identified.
Fig. 5. SEM micrographs showing fracture surfaces of IBA pellets rapidly sintered at: (a) 1020°C and (b) 1050°C.
4.5. Leaching test results

4.5.1. Acid neutralisation capacity (ANC)

ANC data for milled IBA, and IBA sintered at 1050°C is shown in Fig. 6. This indicates that milled and sintered IBA have very similar ANC behaviour except in the acid addition range between 0.4 and 1.4 meq/g where the milled ash gives higher leachate pH values. CaCO₃ is present in the milled IBA as a result of carbonation during ageing, and is reported to provide buffering capacity in the pH range between 5 and 6, due to the formation of Ca²⁺, HCO₃⁻ and OH⁻ ions (Johnson, 1995; Kirby and Rimstidt, 1994; Meima and Comans, 1997). The sintered IBA pellets do not show this effect due to the decomposition of CaCO₃ to CaO and CO₂ that occurs during heating. The CaO is then encapsulated and incorporated into both amorphous and crystalline phases present in the sintered material from which it is extensively leached only under lower pH conditions.

4.5.2. Leaching in distilled water

In addition to the average composition of <8 mm IBA, Table 1 also contains leachate analysis data for milled and sintered IBA after exposure to distilled water leachant (0 meq/g) for 48h. Under these conditions the sintering process significantly reduces the levels of leaching of most metals and particularly Na, K, Al, and Cu. However, the data also indicates increased leaching of Cr, Cd and Zn under these conditions, although this remains at low levels and further data is needed to confirm this effect.
4.5.3. Effect of leachate pH on the release of heavy metals

The effect of leaching under acid leachate conditions (pH 7–2) on the release of heavy metals of particular environmental concern (Cu, Zn, Pb, Cr and Cd) is shown in Fig. 7.
There is very little difference in metal leaching between milled IBA and sintered milled IBA. Previous work has shown that higher temperatures and longer dwell times are needed to significantly reduce metal leaching from sintered IBA (Bethanis et al., 2002a, 2002b). There are however, significant differences in the leaching behaviour of different metals. The fraction of the total metal leached under aggressive acid (leachate pH 3) conditions is considered to be the fraction that is potentially available for leaching in the environment. Cu, Cr and Cd are leached at relatively low levels, with approximately 3 to 8% of the total being potentially leachable. In comparison, ∼33% of the total Zn and ∼42% of the total Pb in the IBA was leached under aggressive acid leachate pH conditions.

5. Discussion

5.1. Technical considerations

Rapid sintering at relatively low temperatures of spherical pellets made from the less than 8mm fraction of IBA produces LWA with properties comparable to commercially available LWA products. Densities (apparent specific gravity) of IBA pellets were typically around 1.55 g/cm³ with water absorption in the range from 10 to 15 wt.%. Individual pellet crushing strengths are comparable, although typically lower than those of Lytag which is manufactured from sintered pulverised fuel ash (PFA).

Production of LWA from milled IBA occurs due to liquid phase sintering resulting from the significant level of glassy material present in milled IBA. The sintering process bonds together the numerous crystalline and refractory phases present in the IBA. XRD data also shows that significant crystalline transformations occur during sintering, and these result in the formation of a number of common rock forming minerals.

Diopside and clinoenstatite were the major minerals formed in sintered IBA, and these belong to the pyroxene group of silicate minerals. They occur as stable phases in almost every type of igneous rock, and they can be found in rocks of widely different compositions. Diopside is a calcium pyroxene, while clinoenstatite is a magnesium-iron pyroxene. Wollastonite also forms in sintered IBA and is a common mineral in metamorphosed limestone and some alkaline igneous rocks. It often occurs with calcite, tremolite, diopside, anorthite and a number of other rare calcium–magnesium silicate minerals (Deer et al., 1992). These mineral phases have also been reported to form in glass–ceramics produced by thermally treating mixes of a number of other municipal and industrial wastes including IBA, incinerator fly ash, coal fired power station fly ash, waste fluorescent glass and electric arc furnace dust (Bocaccini et al., 1995, 1997, 2000; Erol et al., 2000, 2001; Gao and Drummond, 1999; Övecöl et al., 1997; Park and Heo, 2002; Rincón et al., 1999; Romero et al., 1999; Yun et al., 2002).

The effect of sintering milled IBA in the temperature range necessary to form LWA pellets does not significantly reduce leaching of heavy metals. It was expected that heavy metals would be incorporated into amorphous and crystalline phases and therefore not available for leaching. However, the leach test data does not demonstrate any significant difference between leaching from milled and milled/sintered IBA. The fraction of heavy metals in sintered IBA that leach is relatively low under acid conditions. The exceptions are Zn and
Pb. Zn was present at about 3200 mg/Kg in the <8 mm faction of IBA and approximately 33% by weight of this was potentially available for leaching, while for Pb 800 mg/Kg was present, of which approximately 42% was leachable at leachate pH ~3.

### 5.2. Economic considerations

Samples of IBA used in this research were collected from a modern EfW plant that produces approximately 110,000 tonnes of IBA/year. Of this, approximately 50,000 tonnes is expected to be in the less than 8 mm fraction that has been used in this work to manufacture LWA. Current landfill disposal costs in the UK range from £35–£65/tonne depending on the site and location, and this is expected to increase significantly in the next few years. Therefore, landfill disposal of this problematic fraction of IBA represents a significant potential cost.

LWA typically sells in the UK for between £ 40 and £ 180/tonne depending on grade, quantity, application and location. Therefore there is significant scope to cover the processing costs associated with LWA manufacture from IBA involving particle size reduction by milling, pelletising and low temperature rapid sintering. Again, these costs will vary depending on a number of factors including exact processing methods used and availability of by-product energy from waste incineration. However there appears to be potential to produce LWA derived from IBA, and as disposal costs continue to increase and the demand for lightweight concrete products increases, LWA production from IBA will become increasingly viable. The aim of ongoing research is to further optimise processing of IBA to produce low-cost LWA at pilot plant scale, so that this increasingy important waste can be fully exploited as a potential resource.

### 6. Conclusions

(a) The less than 8 mm fraction of IBA has been milled, pelletised and rapidly sintered at temperatures between 1000 and 1050 °C. This produces LWA with densities, water absorption and crushing strengths comparable to commercially available LWA products.

(b) Liquid phase sintering forms the LWA pellets and also causes significant changes in the crystalline phases present. Milled IBA contains quartz (SiO₂) and calcite (CaCO₃) as the major crystalline phases, together with some hematite (Fe₂O₃). IBA sintered at 1030 °C contains reduced levels of quartz together with the common rock forming minerals wollastonite (CaSiO₃), diopside (CaMgSi₂O₆) and clinoenstatite (Mg₂Si₂O₆).

(c) Leaching of heavy metals from IBA under acid conditions is not significantly affected by sintering at the relatively low temperature used to form LWA pellets. Pb and Zn are the major heavy metals available for leaching at pH ~ 3.

(d) LWA production using IBA represents a beneficial and more sustainable reuse for this material than disposal to landfill.

(e) The potential for producing LWA with comparable properties to the commercially available product Lytag using a significant fraction of IBA has been demonstrated. Further work is needed to increase production volumes and further assess bulk LWA
properties, accurately assess production costs and develop IBA derived LWA concrete products.

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