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Sustainable Materialization of Residues from Thermal Processes into Products

Thermal plasma processing in the production of value added products from municipal solid waste (MSW) derived sources

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Abstract

The disposal of Municipal Solid Waste (MSW) is an increasingly difficult challenge for communities. Advanced thermal treatment (ATT) technologies, such as Gasplasma®, have been specifically developed to effectively utilise wastes to generate clean electrical power. An added beneficial feature of this process is the conversion of ash-type materials in the MSW to environmentally-stable products capable of reuse in various applications, closing the recycling loop. This paper discusses this application of Gasplasma®. Also discussed is the use of plasma technology to thermo-chemically treat hazardous Air Pollution Control (APC) residues, derived from the gaseous abatement systems of MSW thermal treatment technologies to produce ceramic products.

Introduction

A major societal challenge that we face lies in the need to develop economic and environmentally acceptable solutions for managing the ever increasing volumes of wastes that are generated worldwide. This must also be considered against the over arching need to find ways of combating the effects of anthropogenic global warming. In this context it is important that the sustainable solutions developed are energy efficient and capable of recovering a high proportion of material values from the waste, thus reducing dependency on primary resources and decreasing the amount of residual materials that are sent for disposal to landfill.

Thermal plasma arcs are characterised by their high temperature and intense, non-ionising radiation. The energy density of the plasma arc is typically 2 orders of magnitude higher than a combustion flame.[1] When applied to the treatment of wastes advantages cited for the technology include:[2] i) high melting and reaction rates leading to compact reactor geometry with rapid start-up/shut-down capability, ii) flexibility in processing a wide range of waste forms, iii) an energy input which is readily controllable and (unlike combustion systems) is independent of the process chemistry and iv) the very low gas volumes associated with plasma processing, which reduces the size, complexity and cost of the downstream gas cleaning equipment.

Thermal plasma processing has also been employed for the treatment of a wide range of hazardous inorganic waste streams including: steel plant residues,[3] MSW incinerator residues,[4,5] asbestos containing materials[6] and aluminium residues.[7] In these applications, plasma arc heating is used to vitrify the waste to form a stable, non-leachable product where heavy metal pollutants are effectively incorporated within a glass network. Although plasma processing is thermally efficient, it utilises expensive electrical power which has often been historically cited as a barrier to its wider adoption. However, this position has changed as a result of escalations in the costs of the integrated landfill disposal benchmark, restriction on the international/regional movement of waste and the increasing stringency of licensing and compliance regulation. The combined factors have positioned plasma technology as an essential component of any future sustainable waste management infrastructure. In this context it is important to emphasise that plasma based recovery solutions are typically more economic than integrated landfill disposal.

The need for more innovative sustainable solutions to the MSW problem are required to ensure more efficient overall use of resources, especially with regards to reducing the power required for vitrifying the waste and lowering the overall environmental impact of the process. One such approach has been adopted by Advanced Plasma Power (APP) which has developed an ATT, primarily for the treatment of household and industrial and commercial (C&I) wastes, which incorporates a plasma processing stage for conditioning the syngas generated from a primary gasification unit and also vitrifies the inorganic (ash) fraction of the feed which may be used as a construction material (see following section).

The plasma vitrified product, derived either from mixed or inorganic wastes may find direct use, for example, as a pipe bedding or unbound aggregate material.[8] However, further processing of this glass may be undertaken to produce high added value engineered materials, which can compete with commercially available products, in architectural or building applications, for example. In this context, one of the most cost effective ways of enhancing the quality of the vitrified products without making major changes to the process is in the production of glass ceramic materials. One specific process route developed by Tetronics, relates to the treatment of Air Pollution Control (APC) residues/fly ash residues obtained from modern Energy from Waste (EfW, see also WtE: Waste-to-Energy) facilities in which the plasma vitrified product may be utilised as the raw material for glass-ceramic production.[9]

This paper describes the plasma vitrification of wastes derived from MSW sources with reference to both the Gasplasma® and Tetronics direct plasma processing technologies. The techniques used in the downstream processing of the glass, to produce high quality ceramic products, are described. Results are also presented regarding the leachability, mechanical and applications testing work that has been undertaken on both the vitrified glass and processed glass-ceramic materials.

Plasma processing/vitrification of MSW derived materials

Market considerations

The total volume of Municipal Solid Waste (MSW) is very considerable and is anticipated to increase in the future. Recent estimates indicate that the global annual amount of MSW produced was between 1.7 and 2.2 billion tonnes.[10] It has been established that there is a clear correlation between economic activity and the volumes of waste produced, where regions of high economic growth produce a corresponding rapid increase in amount of waste generated. For example, in China, the increase in MSW arisings is reported to be between 8-10% per annum.[11] Within the EU 27 countries the amount of MSW waste generated in 2004 was around 266 million tonnes and this is forecast to increase to around 338 million tonnes by 2020.[12]

Against this background, there is a significant and growing amount of residues that are produced from the thermal treatment of waste in MSW energy from waste (EfW) facilities. An especially problematic issue for the EfW industry relates to the management of fly ash and APC residues that are generated in the cleaning of particulate and gaseous emissions as they require some form of physico-chemical or thermal treatment before either disposal to landfill or reuse is permitted. The production rate of APC residue material is around 3.5% of the front end feed rate,[13] which for an estimated EfW treatment fraction of 20% in the EU 27 gives a total production rate of c. 1.86 million tpa (2004 figures). This material is classified as a hazardous waste (absolute entry) under the European Waste Catalogue (19 01 07) on account of its high alkalinity (> pH 12) and other pollutant species, including dioxins and furans, heavy metals and soluble chloride and sulphate salts.

Technological Description and experimental methodology

A schematic of the two-stage thermal Gasplasma® process for advanced thermal treatment of wastes is shown in Figure 1. In the treatment of refuse derived fuel (RDF) the bubbling fluidised bed gasifier (BFBG) is typically operated at a temperature of between 700-850°C, with oxygen and steam being used as the fluidising medium. The gasifier converts the prepared RDF to a raw syngas which, together with the bottom ash from the gasifier, is subsequently treated in a closely coupled plasma converter unit to give two product streams: i) A calcia-alumina-silica rich slag within the compositional range of the anorthite phase field, which upon cooling is both mechanically strong and highly resistant to leaching, and ii) a syngas which (after cooling and acid gas cleaning) can be used for efficient power generation in a gas engine or gas turbine, for conversion to a liquid fuel, or used as a chemical process precursor.

The high energy density of the plasma arc within the converter, which is transferred directly from the plasma device to the molten slag, aids the thermo-chemical processes occurring in both the gas and condensed phases. The intense ultraviolet (UV) light and elevated temperature from the plasma assists the thermal breakdown of the tarry species within the gas space, which would otherwise be highly problematic in the downstream cleaning/power generation stages. Furthermore, the very high heat transfer rates that are attained in the arc impingment zone ensure a high degree of fluidity of the slag so that any solid particle that contacts the melt surface is readily captured and assimilated.

The adoption of a two-stage thermal processing approach used in the Gasplasma® process, enables high energy and carbon conversion efficiencies to be attained in the production of a clean syngas and a vitrified product. Moreover, the majority of the energy input to the thermal process is derived from the controlled oxidation reactions of the solid fuel at the gasifier and this greatly limits the plasma arc electrical power requirement in the converter.

APP has a demonstration Gasplasma® plant in Swindon, UK, with a maximum equivalent capacity of RDF of 100 kg/hr and which has been extensively tested with over 1200 hours of operation. The core principle of the gasifier and the plasma converter described above is applied for both the full scale and the demonstration plant. In the demonstration unit the vitrified slag layer accumulates in the base of the unit and is intermittently tapped whilst on the commercial plant there is continuous removal of the slag from the converter unit.

Tetronics Limited has developed a fully integrated process for the vitrification of APC residues which is capable of handling a wide range of ashes of varying particle size and chemical composition. The APC residue waste is high in calcia and, therefore, silica and alumina additions are made to the feed to provide sufficient network formers to produce a single amorphous glassy phase,[14] these additions are typically made in the form of complementary waste streams. The plasma vitrified material is capable of further materials processing to fabricate enhanced quality ceramic glass

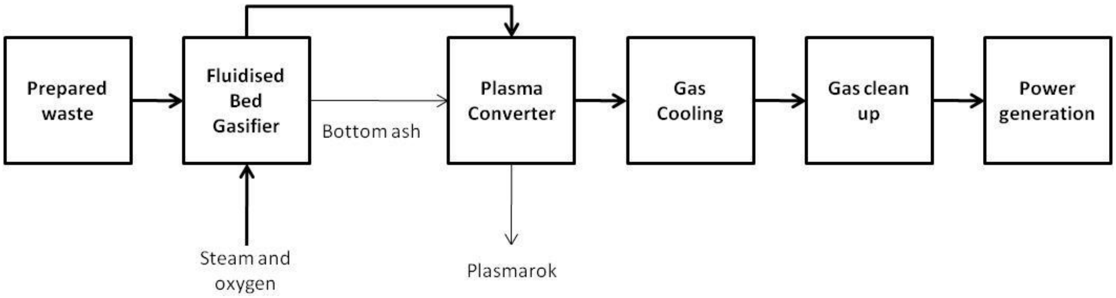

Figure 1: Schematic of the Gasplasma® system (bold lines indicate the waste fuel/syngas flows)

products (see section below). The plasma system consists of a DC hollow graphite cathode electrode located in the centre roof of a furnace whose movements are controlled by a manipulator. An inert plasma gas (Ar or N_2) is injected down the centre of the cathode to produce a stable arc that is transferred to the furnace melt. The return anode electrode consists of conductive elements built into the furnace hearth. The melt temperature is maintained at around 1600°C under a controlled inert gas atmosphere. Remote water cooled elements are employed at the melt line to form a protective frozen layer, ensuring good refractory performance. For commercial operation, typical capacities of ash vitrification facilities using this technology are between 20-30 ktpa.

A demonstration unit for treating APC residues has been comprehensively tested and proved at Tetronics Swindon facility. In a typical experimental run, the plasma furnace is heated up over a period of c. 2.5 h, before introducing the blended feed at a rate of ~80 kg/h for c. 4.5 h. The plasma power is closely controlled to maintain the operating temperature and overcome thermal losses. A layer of untreated feed is maintained on top of the molten slag, where gasification reactions occur. The feed is rapidly assimilated and the molten slag phase is periodically tapped, and cast into ingot moulds. Exhaust gases exiting the unit are treated in a thermal oxidiser unit to fully oxidise residual combustible gas species (CO, H_2). The particulates are removed in a bag house filter and acid gases are removed by the wet scrubber prior to venting to atmosphere.

Materials used in vitrification trials

For the APP Gasplasma® trials, the prepared RDF comes from a number of waste treatment facilities.[15] Table 1 presents the experimentally derived proximate analysis, ultimate analysis and gross calorific value (GCV) of an RDF waste obtained from a UK Mechanical Biological Treatment (MBT) facility and the chemical composition of the vitrified product.

Typical composition data for the APC feed material is shown in Table 2.[16] The physical properties and chemical compositions vary depending on the type of plant and the air pollution control system. The high CaO content is due to excess lime used in the scrubbing process, while the high levels of chloride result mainly from the significant volumes of polyvinyl chloride (PVC) found in MSW.

Table 1: Proximate and ultimate analysis of the RDF material and vitrified slag product for the Gasplasma® testwork

Characteristics	Component
Proximate analysis, %(w/w)	
Fixed carbon	11.6
Volatile matter	64.8
Ash	12.1
Moisture	11.5
Ultimate analysis, %(w/w)	
Carbon	43.0
Hydrogen	5.6
Oxygen	26.6
Nitrogen	0.61
Sulphur	0.25
Chlorine	0.34
GCV, MJ/kg (dry basis)	21.0
Bulk slag analysis, %(w/w)	
Silica	33.3
Calcia	26.6
Alumina	13.8
Iron oxide	15.3
Soda &Potash	5.6
Others	5.4

Characterisation of the vitrified products

The Plasma treatment of the RDF material in the Gasplasma® resulted in a volume reduction of ~800:1 when comparing the vitrified material to the RDF feed. The chemical composition of the vitrified material when treating RDF is given in Table 1 where the primary components are SiO_2.

Plasma treatment of APC residues blended with SiO_2 and Al_2O_3 resulted in significant volume reduction of about 70-75%. The composition of the resulting glass, using XRF analysis, is shown in Table 2.

Table 2: XRF analysis of the as-received and plasma treated APC residues

Chemical species	**As-received APC residues (wt%)**	**APC residue derived glass (wt%)**
Na_2O	3.8	0.4
MgO	0.7	1.2
Al_2O_3	2.2	21.3
SiO_2	4.2	37.7
P_2O_5	0.6	0.35
K_2O	4.2	0.1
CaO	51.9	34.1
TiO_2	0.9	1.3
Mn_3O_4	-	0.2
Cr_2O_3	0.07	< 0.05
Fe_2O_3	0.8	0.9
ZrO_2	0.02	0.05
ZnO	1.3	< 0.5
SrO	0.08	0.06
BaO	0.03	0.06
S	-	0.13
Cl^-	22.3	2.2

XRD analysis gave broad spectra of around 30° indicating that the APC residues and APC-derived glass is amorphous and that crystalline phases have not been formed as shown in Figure 2.

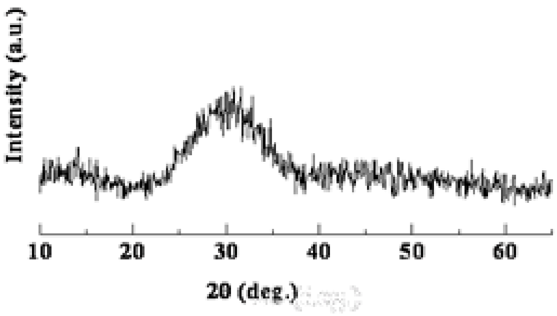

Figure 2- XRD trace showing broad spectra around 2θ of 30°

Ecological compatibility testing of vitrified materials

The results of compliance leaching testing (BS EN 12457-3) of the vitrified material, from the Gasplasma® treatment of RDF, and the APC derived glass are shown in Table 3. The concentrations of heavy metal species in the leachates are seen to be well below the compliance (WAC) limits set for inert landfill for both the materials tested. It is of special note that in the case of the APC residue derived glass, the chloride and sulphate leachate values were only 0.2 and < 50 mg/kg, respectively, whilst the original APC residues typically leaches 140000 to 170000 mg/kg of chloride and 1200-7000 mg/kg of sulphate.[16]

Table 3: Summary of compliance leaching tests (BS EN 12457-3) on vitrified granulated sample at particle size < 4mm. All leachate values are given in mg/kg.

Elements	Gasplasma® RDF derived glass	APC derived glass	Leachable metals/ions in APC residues	Inert Landfill WAC
As	0.025	< 0.007	0.005-4	0.5
Ba	0.040	0.053	10-45	20
Cd	< 0.0055	< 0.0025	< 0.5	0.040
Cr	< 0.02	< 0.016	0.5-2.5	0.5
Cu	0.035	0.076	1.3-3	2
Hg	< 0.0017	0.0031	0.04-0.7	0.01
Mo	< 0.025	0.012	1-4	0.5
Ni	0.010	0.023	0.2-45	0.4
Pb	< 0.03	< 0.007	300-700	0.5
Sb	0.014	0.06	< 0.001-0.02	0.060
Zn	0.120	0.020	40-85	4.00
Cl	< 20	0,2	140000-170000	800
SO_4	< 50	< 50	1200-7000	1000
TDS*	268,0	592,0	-	4000

< Indicates below the lower limit
*Total dissolved solids
L/S=10

Applications testing of plasma vitrified products

A number of aggregates applications tests were performed by a UKAS accredited laboratory on air cooled slag samples obtained from the Gasplasma® process treatment of RDF. The results from these tests were reviewed by an independent consultant to evaluate potential use for the material and to confirm that it complied with the necessary testing standards common to both primary and recovered aggregates.[8] A summary of the tests undertaken and results obtained is given in Table 4. On the basis of this work it was concluded that the material could be readily used for a number of applications including as an unbound aggregate for pipe bedding or road fill sub base material.

Table 4: Results of Applications testing from vitrified product obtained from Gasplasma® testwork

Application test	Purpose of test	Result	Interpretation
Los Angeles test (BS EN 1097-2)	This is a mechanical testing proceedure to demonstrate that the material exhibits acceptable resistance to fragmentation	27%	The material is categorised as LA20 which indicates that there is some minor fragmentation under high loadings, most likely due to the fracturing of the sharp edges on individual particles, whilst the majority of the particles remained intact. The LA coefficient falls well within the range for use as an unbound pipe bedding material where a value of less than 50 is required.
Water soluble sulphate (BS EN 1744-1)	This test measures the level of leachable sulphates in the material that may chemically react with adjacent building materials such as concrete	0%	The vitrified product is categorised as AS0.2 and is virtually inert with respect to sulphate leaching
Magnesium sulphate (BS EN 1367-2)	This test provides a measure of the weathering properties of the aggregate	1%	The materials is categorised as MS18, which indicates that it exhibits extremely high resistance to weathering
Water absorption test (BS EN 1097-6)	This gives a measure as to the materials susceptability to frost heave	0.6%	The material is categorized as WA24 which indicates that the material has low water absorption properties and is not prone to break down under freeze-thaw cycling conditions

Fabrication of higher added value products from plasma vitrified MSW material

General background

The various techniques used in the fabrication of glass-ceramics and other marketable products from vitrified feeds obtained from waste sources are given in review papers by Rawlings, Wu and Boccacicini[17] and Colombo *et al*.[18] The choice of which fabrication technique to employ will be determined by such factors as: its impact on the main front end process, the characteristics of the vitrified feed, the property requirements of the material in the end use application, as well as the obvious economic considerations.

Glass-ceramic products may be fabricated from vitrified feed stocks without requiring major modifications to the process itself. These materials are made by controlled crystallisation of the glass, by creating a high density of crystal nuclei within the bulk of the glass which initiates growth of the crystals to form a fine grained material, which exhibits good mechanical and chemical leach resistant properties, superior to those of the parent glass. Close thermal management of both the nucleation and crystal growth stages is required in order to produce an acceptable and consistent quality of product.

Only certain glass formulations may be used for the production of the glass-ceramics, and therefore glasses which, either crystallise too readily in an unpredictable way, or are very stable and highly resistant to crystallisation, are not suitable. The use of additives to the waste feed material is generally needed in order to produce a glass which falls within an acceptable compositional range for glass-ceramic production. There may also be the need to add nucleating agents such as P_2O_5 and TiO_2 to the feed blend to assist the formation of the crystal nuclei at the nucleation stage of the process.[17]

The different processing routes that are used to produce glass ceramic materials from waste materials may be summarised as follows:

- Conventional nucleation and growth: The classical two stage method of producing glass ceramic materials is by crystallising the parent glass by a low temperature nucleation stage, to produce a high density of nuclei, followed by higher temperature secondary stage to promote rapid crystal growth at the previously formed nucleation points. This process occurs by heating the glass from room temperature. In cases where there is significant overlap between the nucleation and crystal growth rate curves it is possible to produce the glass ceramic material in a single heat treatment stage. This is commonly referred to as the modified conventional single stage method, which reduces the complexity, time and cost of production.

- Peturgic method: This refers to a technique whereby certain glass-ceramics may be produced by controlled slow cooling of the parent glass from the molten state. The major benefit of this technique is that it eliminates the need for an expensive thermal treatment stage and also simplifies the overall production of the glass-ceramic material.
- Powder sintering: Glass ceramic compacts are formed by simultaneous sintering and crystallisation of a ground glass powder at an optimum temperature. This temperature is typically lower than that associated with the previous two methods due to the prevalence of surface nucleation. Potential benefits cited for this technique are that the process may be carried out at a dedicated ceramic production facility which is remote from the waste treatment site. In this case the vitrified waste may be a small fraction of the total feed blend, allowing for a wider materials specification to be accepted for this material. Furthermore, the large amount of surface nucleation sites ensures a large number of fine crystals without the need for the admixing of nucleating agents.

There are also a number of alternate manufacturing methods, which have been used in the production of high value products from glass, including glass fibres, geopolymer, foams and composites.[18] The following section briefly describes the investigative work conducted at Imperial College, London (ICL) and reported by Rani *et al.* [9]on the properties of glass ceramics tiles formed by the powder sintering of Tetronics' plasma technology derived APC residue glass and how these compared against commercial selected ceramic tile materials.

Production of sintered glass ceramics from plasma vitrified material

Plasma vitrification

The APC residue was obtained from a major UK EfW incinerator facility. The blended feed material used in the plasma vitrification trials consisted: 69.8 wt% APC residues, 21.9 wt% SiO_2 and 8.3 wt% Al_2O_3. This gave a normalised bulk solid oxide input composition in the anorthite region of the Al_2O_3–SiO_2–CaO phase diagram. The molten APC residue-derived glass from the plasma treatment was quenched into water to produce a fritted sample with particle sizes of c. 5 mm. The chemical compositions of as received and the plasma treated APC residues are given in Table 2.

Tile production

The plasma vitrified APC material derived from APC residue and cullet glass (CG) were separately milled to < 250 µm. Three different mix compositions were prepared: (a) 100% APC (b) 50% APC and 50% cullet glass (50PVM:50CG) (c) 100% cullet glass (100CG).

A bentonite (5 wt%) binder was added to each batch and the powder samples were thoroughly mixed in a laboratory mixer with ~20 wt% water added to produce granulated spheres which were seived through a 1.18 mm mesh to give a free flowing powder. A rectangular die was uniformly filled with the powder and pressed at 250 kg/cm to form a green tile. These were sintered at temperatures in the range of 700°C and 1100°C, for 1hour after being heated at a rate of 10°C per min.

Tile Properties

A summary of typical measured properties of the APC derived glass ceramic tiles as compared with typical values of commercial ceramic tiles are given in Table 5. The key findings from the study were as follows:

Density and water absorption data

The bulk density of the 100APC sintered samples was 2.4g/cm^3 which was comparable to the commercial porcelain tiles. The bulk density of the 50:50 APC:CG gave a maximum value of 2.15 g/cm^3 attained at a sintering temperature of 900^{o}C. The 100CG samples deformed at higher temperatures and had a maximum density of 1.85g/cm^3. In general it was observed that the sintering temperatures under which glass-ceramics were formed were significantly lower than those required for commercial tile manufacture.

The water absorption figure provides an index as to the volume of open pore structure in the sintered bodies. It was observed that for the 100APC samples that the water absorption was comparable to the floor tiles and lower than the porcelain tiles. It was stated that further densification of the powder compact could be achieved by reducing the particulate size of the precursor powder.

Strength and hardness measurement

The flexural strength and hardness values for selected trials are given in Table 5. It was observed that the flexural strength of the 100 APC tiles was around 60 MPa, which was higher than commercial tiles and did not vary significantly with sintering temperature over the range 900-1100^{o}C.

The Vickers hardness values had an average value of 5.4 GPa, which is higher than values reported for the porcelain tiles.

Table 5: Measured properties of APC residues derived glass ceramic tiles. Typical values of commercial and reported tiles[9]

100 APC – 100 wt% APC residues derived glass
50:50-APC:CG – 50 wt% and 50 wt% mixed APC derived and cullet glass

Properties	**100APC**			**50:50 APC:CG**	**Floor tile standard**[a]	**Wall tile standard**[a]	**Multi purpose floor tiles**[a]	**Multi purpose wall tiles**[a]	**Wall tiles**[b]	**Floor tiles**[b]	**Vitrified tile**[b]	**Porcelain tiles**[c]
Firing temperature (°C)	900	1000	1100	900	1200	1170	1200	1170	1060	1175	1175	1220
Density (g/cm^3)	2.4	2.4	2.4	2.2	-	-	-	-	-	-	-	2.42
Water absorption (%)	5.3	5.6	5.7	6	2.0	14.5	1.2	11.0	14.2	4.0	0.2	0.08
Linear shrinkage (%)	4.0	4.45	4.7	2.7	6.0	-0.2	5.0	0.6	0.9	4.0	5.8	-
Young's modulus (GPa)	95.5 ± 4	90.3 ± 3	86.6 ± 3	62.1 ± 6	-	-	-	-	-	38.7	-	-
Flexural strength (MPa)	60.6 ± 2	61.1 ± 1	58.3 ± 1	31.2 ± 7	48.32	27.33	46.53	30.0	1.62	-	60.0	61.0
Hardness (Hv) (GPa)	5.5 ± 0.1	-	4.2 ± 0.9	4.5 ± 0.9	-	-	-	-	-		-	7.3

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[a] Kara *et al.*,[19] *JECS* (2006) (negative sign means expansion)

[b] Ghosh *et al.*,[20] *Industrial ceramics* (2006)
(floor and wall tile – 90: 10 % pyrophyllite: clay; synthetic vitrified tile – 50 : 50% pyrophyllite: clay)

[c] Carbajal *et al.*,[21] *JECS* (2007)

Conclusion

Conclusions derived from this work are:

- Municipal solid waste can be converted to clean power, whilst simultaneously converting ash-type components in the MSW into a vitrified product of low leaching potential.
- The vitrified product from MSW conversion to power, has properties suitable for use in aggregate applications, with low mechanical fragmentation, low sulphate leaching, good weathering properties, and good frost resistance.
- The vitrified product from MSW conversion to power can also be considered for use in production of added value materials including glass-ceramic products.
- Air Pollution Control (APC) Residues from gas cleaning processes can be vitrified using plasma technology to eliminate the hazardous nature of the residues and yield a saleable glassy product. Further enhancement of the material properties can be achieved utilising glass-ceramic sintering techniques. The properties of APC derived glass ceramic tiles compared favourably with typical values of commercial ceramic tiles.

Acknowledgements

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