

CARBONATION OF MSWI APCr FOR RE-USE AS A LIGHT-WEIGHT AGGREGATE

P.J.Gunning¹, [C.D.Hills², P.J.Carey³, S.Greig⁴ and M.Schilling⁵]

¹Carbon8 Systems Ltd, Medway Enterprise Hub, Chatham Maritime, United Kingdom, +44 (0)2083319848, info@c8s.co.uk

²Centre for Contaminated Land Remediation, University of Greenwich, Chatham Maritime, United Kingdom, +44(0)2083319820, c.d.hills@gre.ac.uk

³Carbon8 Aggregates Ltd, Medway Enterprise Hub, Chatham Maritime, United Kingdom, +44 (0)2083319848, paulacarey@c8s.co.uk

⁴Carbon8 Aggregates Ltd, Medway Enterprise Hub, Chatham Maritime, United Kingdom, +44 (0)2083319848, stevegrieg@c8s.co.uk

⁵Carl von Ossietzky Universität Oldenburg, Germany, macourielle@aol.de

Abstract –The incineration of municipal solid waste (MSW) is a more sustainable alternative to landfilling. Cleaning of flue gases from the incineration of MSW generates air pollution control residues (APCr). Carbon8 Systems, a spinout company from the University of Greenwich, UK has developed a treatment for APCr using accelerated carbonation technology (ACT). Accelerated carbonation enables carbonate-based solidification of an ash (or other suitable waste) using carbon dioxide gas. The solidified and stabilised products can be formed as pellets that are suitable for use as aggregate as a component in the manufacture of concrete building blocks. The result of this work is laying foundations for the construction of the world's first full-scale commercial accelerated carbonation plant in the UK, with a capacity to produce 36,000 tonnes of APCr-derived aggregate annually.

Keywords: MSWI, APC, Accelerated Carbonation, Recycled Aggregate

INTRODUCTION

The UK generated 35.1 million tonnes of municipal waste in 2006. Around 64% of this was disposed to landfill. As landfill usage is costly and is being discouraged, recycling and incineration are attractive alternative management strategies. Nearly 5.2 million tonnes of waste was incinerated in 2007; an increase of six per cent on 2006 [1]. Currently 33 incinerators have a combined permitted capacity of 8.6 million tonnes, but the planned construction of as many as 80 new incinerators will provide a further 12 million tonnes of burning capacity for the UK [1].

Incinerator flue gasses are acidic, and may contain significant trace levels of dioxins and volatile metals. Lime and carbon are usually added to flue stack to neutralise the pH and remove these pollutants. The resulting dust is collected in bag filters before the cleaned flue gas is released to the atmosphere [4]. APCr's are classified as hazardous waste in the European Waste Catalogue, primarily because of their very high pH [5]. Around 128,000 tonnes of APCr is currently generated in the UK each year, rising by a further 40,000 tonnes in the near future [2,3].

Most of the APCr's generated are disposed in either specialized hazardous landfill facilities or placed in underground storage. Facilities for hazardous waste disposal are tightly regulated, and few merchant sites exist in the UK that can take APCr [6]. The properties of APCr often exceed the acceptance criteria for hazardous landfill, and can only be disposed at selected sites, which have received a derogation permit. Hazardous waste is subject to the current active rate landfill tax of £56 per tonne, increasing to £64 in April 2012, and by £8 each year until 2014 [7].

New methods of treating APCr are being developed, which include washing the dusts with water to remove contaminants [8], chemical treatments to that induce dissolution, absorption, or precipitation of contaminants. Thermal treatments including vitrification into a stable glass-like material have been explored [9]. A novel alternative is to solidify and stabilise the APCr using carbonation, whilst at the same time sequestering this harmful greenhouse gas as solid carbonate salts.

Carbonation is a naturally occurring process, which can be accelerated by exposing reactive materials to a higher concentration of carbon dioxide gas than that in the atmosphere. Many wastes are susceptible to carbonation, including those derived from industrial thermal processes, such as incineration residues [10,11,12,13,14,15].

The carbonation process involves a 'series' of chemical reactions [16]. Firstly, carbon dioxide gas undergoes hydration in water, and then reacts with calcium ions (dissolved from the reactive material) to form solid calcium carbonate. The formation of calcium carbonate has the effect of reducing the alkaline pH of the material, achieving minimum solubility of many heavy metal contaminants [17,18]. As calcium carbonate is more voluminous than the reactant minerals, it infills pore space, physically encapsulating contaminants and solidifies the product [19,20,21].

The carbonation reaction can be managed and a process to create hardened pelletised materials from wastes using waste carbon dioxide gas [22]. The process enables fine waste powders to be formed into aggregates with resultant properties which render them suitable for use as a construction aggregate.

EARLY DEVELOPMENT

The development of the carbonation process has extended over ten years, beginning with a lengthy phase of laboratory-based research and development, which demonstrated the production of pelletised materials from a variety of wastes [23,24]. By developing ways of managing the materials and the carbonation process, aggregates with properties comparable to commercially available artificial aggregates, could be produced. The wastes investigated were obtained from a number of industrial processes and included:

- Bauxite: a by-product of the aluminum manufacturing process
- Biomass ash: derived from burning biomass for energy from waste
- Clinical Ash: from the incineration of hazardous medical materials
- MSWI Ash: municipal waste incineration; coarse bottom ash and air pollution control residues
- Paper Ash: from the incineration of paper de-inking and wastewater sludge
- Steel Slag: residues from steel manufacture

A pilot-scale trial followed at a waste treatment facility to demonstrate the process operating at a rate of 0.25 tonnes/aggregate per hour. The aggregates manufactured were used to manufacture concrete construction blocks, as one potential end use of the aggregated product.

The results of the pilot-scale trial facilitated private investment for the construction of a temporary full-scale demonstration plant that produced over two hundred tonnes of aggregate, at a rate of up to 5 tonnes per hour [25]. During the trial both the aggregate and blocks were subjected to rigorous testing by an independent, accredited laboratory.

DEVELOPMENT OF THE FULL SCALE PLANT

As a result of the work outlined above, a full-scale production plant is planned in the UK with the capacity to produce 36,000 tonnes of aggregates a year. The work carried out to date should provide the basis for a 'world-first' production plant. This and the properties of the aggregate and concrete blocks produced hitherto are discussed further.

The production of aggregates by the accelerated carbonation process is shown in figure 1, with an illustration of the plant layout shown in figure 2.

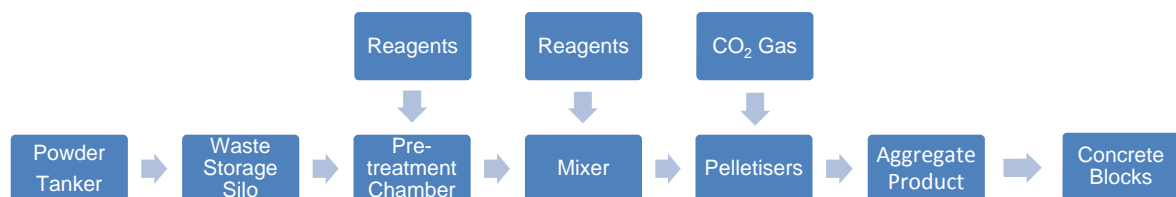


Figure 1: Flowchart of the full-scale demonstration process

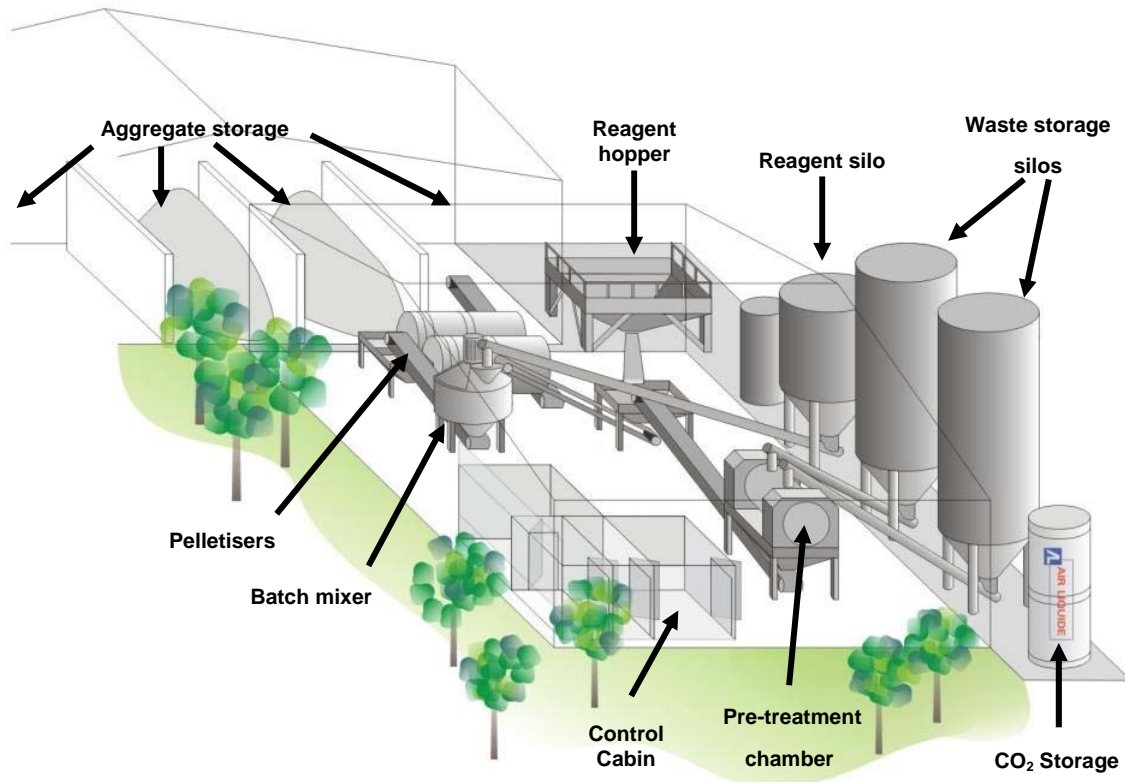


Figure 2: Illustration of the completed manufacturing plant

Silos will be used to store the APCr, but due to the lack of a local point source of CO₂, a large capacity liquid gas tank will be necessary for use in the process. The APCr will undergo treatment and pass through a carbonation step. The formed aggregate will then conveyed to temporary storage bays each holding one day's production. Regular checks on the quality of the incoming APCr and outgoing aggregate product will be necessary to ensure that the aggregate meets the required specification set out in the regulatory agencies compliance certification. The plant is anticipated to be under construction in early 2012, and will receive APCr from incinerators located in the south of England. Over 36,000 tonnes of aggregates product is planned during the plant's first year of operation, and be used by an independent block maker, in the production of medium-weight concrete blocks. Part of the anticipated plant during fabrication is shown in figure 3.



Figure 3: Aggregate manufacturing plant during fabrication

PRODUCT TESTING

Quality standards for the production quality aggregate and the final concrete block products have been agreed as part of the regulatory approvals process, supported by rigorous testing by independent accredited laboratories.

British Standard methods were used to evaluate the pilot-scale production aggregate and manufactured blocks. Aggregate mechanical properties were tested according to BS EN 1097 and BS EN 13055, chemical properties according to BS EN 1744, and block properties according to BS EN 772 [26,27,28,29].

Aggregate Properties

The manufactured aggregate is light grey in colour and forms rounded and sub-rounded pellets less than 15mm in diameter (see figure 4), and has been designed for use in concrete blocks. The physical and chemical properties of the aggregate are shown in table 1. Included are regulatory limit values where applicable, and the properties of another commercially available material, LECA (Lightweight Expanded Clay Aggregate), for comparison. Carbonated aggregate containing APCr is classified as lightweight aggregate, as its bulk density does not exceed 1200 kilograms per cubic metre (kg/m³) [27]. LECA is a sintered material with a bloated structure, which accounts for its lower bulk density.

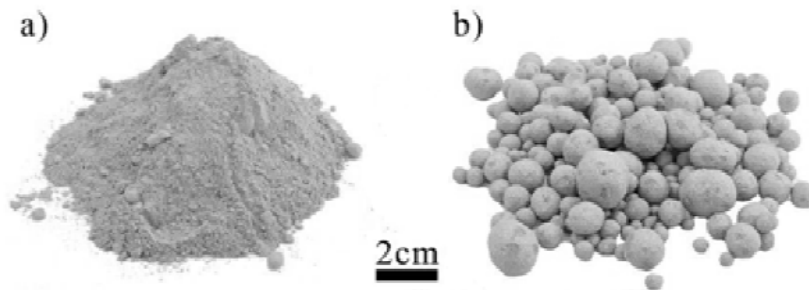


Figure 4a) Untreated APCr, b) APCr aggregate

Crushing resistance is a specific test for lightweight aggregate. The value, stated in newtons per millimetre squared (N/mm²), is the force required to achieve a set amount of compression of a bulk sample of aggregate in a hydraulic press. The value obtained for the carbonated APCr aggregate was an order of magnitude higher than that of sintered aggregate. No limit values exist for crushing resistance, except that the results must be declared, and the aggregate must be of suitable quality to create the required end product.

Table 1. Physical and chemical properties of APCr aggregate

	Grading, mm	Bulk Density, kg/m ³	Crushing Resistance, N/mm ²	Water Absorption, %	Soluble Chloride, %	Total Sulfur, %	Acid Soluble Sulfate, %
APCr Aggregate	15-5	910	11.05	19.6	1.56	0.91	0.2
	<5	1143					
LECA	10	330	1.23	20	0.04	0.32	0.5
Limit Value	N/A ^a	1,200 ^a	N/A ^a	N/A ^a	0.2 ^b	1 ^c	1 ^d

^a According to BS EN 13055-1²⁷, ^b According to BS EN 206 based upon the total weight of cement in the final concrete product³⁰, ^c According to BS EN 13139 based upon the total weight of the final product³¹, ^d According to BS 8500³²

The levels of sulfate and chloride levels are set for the masonry or concrete unit in which the aggregate is used, rather than the aggregate itself. Values are expressed as either a percentage of the total weight of the block (or as a proportion of the weight of the cement used in the block formulation). The blocks were manufactured using a mixture containing no more than 30% by weight

of the carbonated APCr aggregate. Accordingly, the sulfate and chloride levels were well within the limits applicable to the finished product.

Leaching of the carbonated APCr aggregate was determined in accordance with the Waste Acceptance Criteria (WAC) using BS EN 12457-2, which is a single stage dynamic water extraction procedure³³. Untreated ACPr's may leach heavy metals exceeding the regulatory limits for hazardous waste, which include lead, zinc and copper. After processing the MSWI APCr into aggregate, the leachate levels are substantially reduced. The specification given in table 2 shows the levels for key contaminants. In the final block product, the APCr aggregate will form part of the solid monolith and under the prescribed service environment (any) leaching will be further substantially reduced.

Table 2. Leaching values of APCr aggregate

Leaching (mg/kg)													Total Dioxins (ng/kg)
Antimony	Arsenic	Barium	Cadmium	Chloride (x10 ³)	Chromium	Copper	Lead	Molybdenum	Nickel	Selenium	Sulfate (x10 ³)	Zinc	
0.06	0.5	20	0.04	75	1.5	0.15	0.5	1	0.4	0.1	5	3.5	600

The carbon footprint of the aggregate has been initially calculated using CCalc software. Although, carbon is generated during processing, it is balanced by the waste carbon dioxide used to effect carbonation. This carbon dioxide is permanently sequestered in the aggregate and laboratory-based calculations indicate the aggregate is carbon negative. There is potential to realise further carbon savings, which will make the aggregate substantially carbon negative.

Carbonated Aggregate Concrete Block Properties

Carbonated APCr aggregate produced at pilot-scale was used to manufacture heavy-weight (nominal gross density 1990kg/m³), and medium-weight (1350-1500kg/m³) concrete blocks. Their production can be seen in figure 5. A variety of materials, including natural aggregate, and industrial by-products are normally incorporated into the mix formulation, and carbonated APCr aggregate was used as a replacement for these materials. Table 4 shows example formulations used to manufacture the blocks, together with selected key properties.

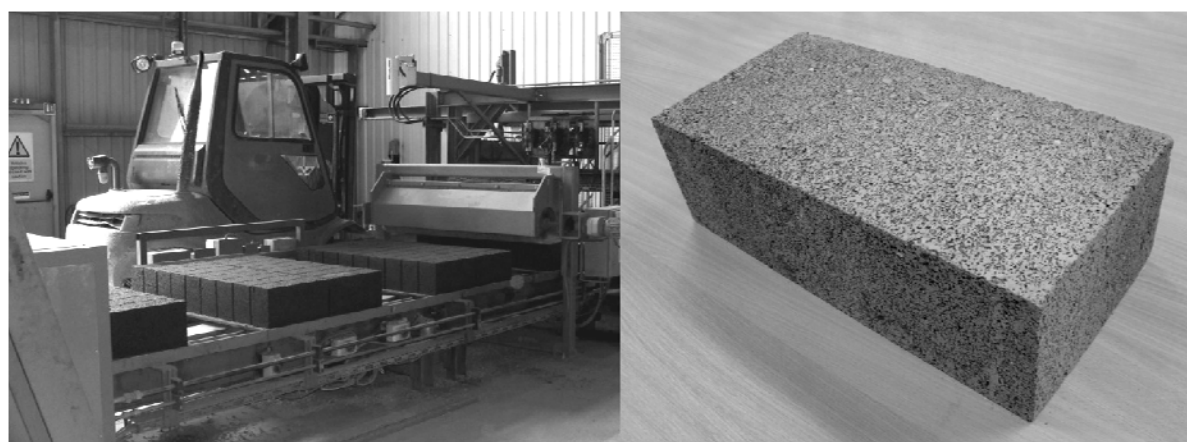


Figure 5: APCr aggregate concrete blocks during production (left), finished product (right)

The carbonated APCr aggregate reduced medium-weight block density and increased block strength. The heavy-weight blocks showed enhanced strength at 7 days, and were 10% stronger when mature, at 28 days. By increasing the addition of carbonated APCr aggregate to 30% in medium-weight blocks, the gross density decreased by 8%, whilst strength was increased by approximately 8%, despite the reduction in cement content of nearly 30%.

Table 3. Physical properties of APCr aggregate concrete and control blocks

	Control Heavy-weight	Heavy-weight 10% APCr	Control Medium-weight	Medium-weight 10% APCr	Medium-weight 20% APCr	Medium-weight 30% APCr
Cement (kg)	260	275	280	229	224	199
Filler* (kg)	4400	4400	2348	2486	2512	2146
Dry Weight (kg)	19.0	18.9	14.4	14.0	13.7	13.3
Density (kg/m ³)	1980	1990	1470	1440	1380	1350
Strength 7 days (MPa)	11.9	12.4	7.9	8.0	8.8	8.5
Strength 28 days (MPa)	16.8	18.4	8.6	9.3	9.5	9.6
Transverse Failure (kN)	8.2	8.6	5.8	6.2	5.2	5.4

*including the APCr aggregate

SUMMARY

A process for the manufacture of artificial aggregate from MSWI ARCr has been trialled at pilot-scale. The technology uses waste carbon dioxide gas to solidify the ash into a monolithic product that has potential to be used as an aggregate in construction. The carbonated aggregate has the physical and mechanical properties that designate it as a lightweight aggregate.

In the carbonation process, the risks associated with the APCr are managed. Carbonated ARCr aggregates are predicted to be carbon negative, containing carbon dioxide gas that is permanently bound in the solidified product.

The development of the process involved laboratory-scale research and development, leading to pilot-scale trials, prior to a production-scale demonstration of the manufacture of 3000 blocks. The results of the development program enabled regulatory approval to be obtained, enabling the basis for the construction of the first commercial plant of its kind in 2012. The planned capacity of the commercial plant is 36,000 tonnes of aggregate in its first year of operation, for use in the manufacture concrete construction blocks.

The incorporation of carbonated aggregate has been shown to lower the density of concrete blocks whilst improving manually handling and reducing transport costs. Increasing the proportion of aggregate used also raises the compressive strength of blocks, whilst allowing the cement content to be lowered by as much as 30%.

The first UK production facility is planned during 2012, it is anticipated that carbonated aggregates will be routinely used in the manufacture of concrete construction blocks.

REFERENCES

- [1] Environment Agency. "Hazardous waste data: 2007". <http://www.environment-agency.gov.uk/> Accessed 02.01.2012.
- [2] Lampris, C., Stegemann, J.A. Cheeseman, C.R. "Solidification/stabilisation of air pollution control residues using Portland cement: Physical properties and chloride leaching". *Waste Management*, 29, 3, 2009, 1067-1075.
- [3] Rani, A., Boccaccini, A.R., Deegan, D., Cheeseman, C.R. "Air pollution control residues from waste incineration: Current UK situation and assessment of alternative technologies", *Waste Management*, 28, 2008, 2279-2292.
- [4] Macleod, C., Duarte-Davidson, R., Fisher, B. Ng, B., Willey, D., Shi, J.P., Martin, I., Drew, G., Pollard, S. "Modeling human exposures to air pollution control (APC) residues released from landfills in England and Wales" *Environment International*, 32, 4, 2006, 500-509.
- [5] Astrup, T., Mosbæk, H., Christensen, T.H. "Assessment of long-term leaching from waste incineration air-pollution-control residues", *Waste Management*, 26, 2006, 803-814.
- [6] Environment Agency. "Treatment and disposal of hazardous waste". <http://www.environment-agency.gov.uk/>. Accessed 02.01.2012.

- [7] HM Revenue & Customs. "A general guide to landfill tax". <http://www.hmrc.gov.uk/>. Accessed 05.01.2012
- [8] Chimenos, J.M, Fernandez, A.I., Cervantes, A., Miralles, L., Fernandez, M.A., Espiell, F. "Optimizing the APC residue washing process to minimize the release of chloride and heavy metals" *Waste Management*, 25, 2005, 686–693
- [9] Amutha Rani, D., Gomez, E., Boccaccini, A.R., Hao, L., Deegan, D., Cheeseman, C.R. "Plasma treatment of air pollution control residues" *Waste Management*, 28, 2008, 1254–1262
- [10] Diener, S., Andreas, L., Herrmann, I., Ecke, H. and Lagerkvist, A. "Accelerated carbonation of steel slags in a landfill cover construction", *Waste Management*, 30, 1, 2010, 132-139.
- [11] Huntzinger, D. N., Gierke, J. S., Sutter, L. L., Kawatra, S. K., Eisele T. C. "Mineral carbonation for carbon sequestration in cement kiln dust from waste piles" *Journal of Hazardous Materials*, 168, 1, 2009, 31-37
- [12] R. Baciocchi, G. Costa, E. Lategano, C. Marini, A. Poletini, R. Pomi, P. Postorino, and S. Rocca, "Accelerated carbonation of different size fractions of bottom ash from RDF incineration", *Waste Management*, 30, 7, 2010, 1310-1317
- [13] Chen, Q., Johnson, D.C., Zhu, L., Yuan, M., Hills C.D., "Accelerated carbonation and leaching behavior of the slag from iron and steel making industry", *Journal of University of Science and Technology Beijing, Mineral, Metallurgy, Material*, 14, 4, 2007, 297-301
- [14] Arickx, S., Van Gerven, T., Vandecasteele, C. "Accelerated carbonation for treatment of MSWI bottom ash" *Journal of Hazardous Materials*, B137, 2006, 235-243
- [15] Johnson, D.C. "Accelerated carbonation of waste calcium silicate materials", Sci. Lecture Series, Society of Chemical Industry, 2000.
- [16] A.Maries, "Measurement of gas consumption during accelerated carbonation of Portland cement mortar", *Proceedings of the 2nd International Conference on Accelerated Carbonation for Environmental and Materials Engineering*, Rome, 1-3 October 2008
- [17] Li, X., Hills, C.D., Carey, P.J., Simons, S. "Making aggregates from waste and carbon dioxide", *Proceedings of the international 23rd cement and concrete science conference, University of Leeds*, 2003.
- [18] Van Ginneken, L., Dutré, V., Adriansens, W., Weyten, H. "Effect of liquid and supercritical carbon dioxide treatments on the leaching performance of a cement-stabilised waste form" *Journal of Supercritical Fluids*, 30, 2004, 175-188
- [19] Rendek, E., Ducom, G., Germain, P. "Carbon dioxide sequestration in municipal solid waste incinerator (mswi) bottom ash" *Journal of Hazardous Materials*, 128, 2006, 73-79
- [20] Johannesson, B., Utgenannt, P. "Microstructural changes caused by carbonation of cement mortar" *Cement and Concrete Research*, 31, 2001, 925-931
- [21] Lange, L.C., Hills, C.D., Poole, A.B. "The effect of accelerated carbonation on the properties of cement-solidified waste forms", *Waste Management*, 16, 1996, 757-763
- [22] Hills, C.D., Carey, P.J. 2007. WIPO Patent WO/2007/096671A1. Production of Secondary Aggregates. World Intellectual Property Organisation.
- [23] Padfield, A.M., Carey, P. J., Hills, C. D., Poole, A. B. 2004. Reuse of quarry fines in production of secondary aggregates. *Engineering Sustainability*, 157, 149-154.
- [24] Gunning, P.J., Hills, C.D., Carey, P.J. "Production of lightweight aggregate from industrial waste and carbon dioxide", *Waste Management*, 29, 2009, 2722-2728
- [25] Gunning, P.J., Hills, C.D. and Carey, P.J. and Antemir, A. "Novel approaches to the valorisation of ashes using aggregation by carbonation". *Proceedings of the 2nd International Slag Valorisation Symposium, Leuven, Belgium*, 18-20 April 2011.
- [26] British Standard Institute, "BS EN 1097: Tests for mechanical and physical properties of aggregates", 1996
- [27] British Standard Institute, "BS EN 13055: Lightweight aggregates – Part 1: Lightweight aggregates for concrete, mortar and grout", 2002
- [28] British Standard Institute, "BS EN 1744: Tests for the chemical properties of aggregates", 1998
- [29] British Standard Institute, "BS EN 772: Methods of test for masonry units", 2000
- [30] British Standard Institute, "BS EN 206-1: Concrete", 2000
- [31] British Standard Institute, "BS EN 13139: Aggregates for mortar", 2002
- [32] British Standard Institute. "BS 8500-1: Concrete – complimentary British Standard to BS EN 206-1", 2006
- [33] British Standard Institute, "BS EN 12457: Characterisation of waste. Leaching. Compliance test for leaching of granular waste materials and sludges", 2002