SAPICO2: Production of Sustainable Construction Aggregates through Cementation with Carbon Dioxide Peter GUNNING¹, Robert GUNNING², Alan MARIES³, Colin HILLS^{2*}

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Abstract

The EU-funded project, Sustainable Aggregate Production with Imbibed Carbon Dioxide (SAPICO2) is an INTERREG IVa Channel Programme examining the development of eco-construction materials made from various carbonate-able wastes residues normally disposed to landfill. The partnership involves the University of Greenwich (UoG), the University of Picardie Jules Verne (UPJV), and Carbon8 Systems (a UoG spin-out company). The project applies accelerated carbonation technology (ACT) to facilitate waste valorisation in the production of manufactured carbonated materials that have re-use potential. SAPICO2 has investigated more than 100 wastes originating in NW France and the SE UK (Channel Region), with a view to explore new ways in which carbonation can be applied beneficially. Wastes have been collected and characterised for their chemical and physical properties, very importantly for their ability to react/combine with CO₂ gas. It is shown that with careful control of a carbonation process reaction conditions it is possible to treat waste to give it value and re-use potential and thus, divert it from landfill into the materials supply chain. Such an outcome meets the needs of the developing European 'circular economy'. The final part of the SAPICO2 project involved the production of bulk samples of carbonated materials for product testing/evaluation in France.

Keywords: aggregate; recycling; waste; CCS; EU

1 Introduction

The SAPICO2 (Sustainable Aggregate Product with Imbibed CO2) project is a collaboration between the University of Greenwich (UK), the University of Picardie Jules Verne (France), and Carbon8 Systems (a spinout company from the University of Greenwich) (Project SAPICO2, 2015). SAPICO2 is a European Regional Development Fund project, supported by the Interreg's Manche (Channel) programme (Interreg 4A, 2015).

The SAPICO2 project aims to develop new carbonation technology for combining gaseous CO_2 with solid waste to manufacture carbonate-cemented eco-construction materials (including aggregates) and introduce this to the French Channel region (The CO_2 Forum, 2015).

Many wastes are susceptible to carbonation, particularly those derived from industrial thermal processes. The carbonation process involves reaction of carbon dioxide gas with primarily calcium and magnesium minerals from the reactive material, to form solid calcium carbonate (limestone). This has the effect of lowering the pH of the material, reducing the solubility of many heavy metal contaminants. The growth of calcium carbonate crystals results in infilling of the pore space and acts as a binding cement, physically encapsulating contaminants and solidifying the material. The process enables fine-grained waste powders to be formed into aggregates with properties which render them suitable for use as a construction aggregates. Based upon available data (between 2007 and 2011), it is possible to sequester up to 1Gtonne of CO_2 by carbonating six alkaline residues found widespread around the world (Gunning *et al.*, 2014).

The first phase of the project involved identifying available waste streams in France and in the UK for use with carbonation treatment. By gathering samples of these wastes, a laboratory assessment of their chemical and physical properties was made, including their capacity to react with CO_2 . In the second phase, those materials reactive with CO_2 were developed into prototype aggregates for preliminary testing. In the final phase of the project, the prototype aggregates meeting the agreed specification were developed further through a series of pilot scale trials to produce bulk quantities for rigorous end-use testing.

The UK project partners have undertook consultations with the Environment Agency (EA), along with the Building Research Establishment, the Quarry Products Association, British Cement Association and the UK Quality Ash Association have taken place. The French team have liaised with the French Environment and Energy Management Agency (ADEME), through their Waste and Soils Management Office and the Picardie Regional and Departmental Councils, as well as with private waste management/processing organisations.

SAPICO2 is now in its third and final phase. Bulk samples of new products have been manufactured using paper ash and municipal waste incinerator residues from the UK, and using biomass ashes and quarry fines from France. In addition to this, plans are in place to conduct a proof-of-concept demonstration to manufacture 1-2 tonnes of biomass ash aggregate for end-use trials in conjunction with a construction block manufacturer in France. This will be a key step in obtaining validation of the aggregate as a product.

Currently, an equivalent to the UK's 'end-of-waste' process does not exist in France. However, it is hoped that this project (and others like it) will motivate a change of approach and that a route to facilitate waste de-classification is opened up in France.

2 Wastes Examined

The European Waste Catalogue has been used to classify wastes in this project. The wastes suited to carbonation processing are listed in chapter 10 (inorganic wastes from thermal processes), and chapter 19 (wastes from treatment facilities, off-site waste water treatment plants and the water industry) (European Commission, 2015).

More than 100 waste samples were obtained from across the UK and France, and were grouped into 13 categories (Table 1).

The ability to predict the carbon dioxide reactivity is important for evaluating sequestration potential. If this can achieved using basic, easily obtainable data, an assessment of a material can be made without the need for laboratory testing. Steinour (1959) devised a formula to predict the maximum theoretical carbon uptake of cements based upon the concentration of calcium, sulphur, sodium and potassium (see eq. 1).

$$CO_2(\%) = 0.785(CaO - 0.7SO_3) + 1.09Na_2O + 0.93K_2O$$
(1)

This formula has been applied to the waste samples obtained, after determining their bulk chemical composition by X-ray fluorescence (XRF). The actual carbon capacities were then determined using an accelerated carbonation technique using a pressurized reaction chamber (Gunning *et al.*, 2010).

The results yielded a valuable insight into the variability of the chemistry of wastes. The compositions of the relevant constituents (expressed as oxides) are shown in figure 1. Concerning; calcium (CaO), potassium (K_2O), sodium (Na_2O), and sulphur (SO_3), the variability within each group can be appreciated. This reflects variations in the raw materials used, operating conditions, plant age, and waste treatment methods.

Table 1. Residues examined

No.	Waste Group	Source			
1	Biomass Ash - Bottom Ash (BBA)	Heavy material remaining in the grate after combustion			
2	Biomass Ash - Fly Ash (BFA)	Lighter material carried from the combustion zone in the flue gasses before removal			
3	Cement Kiln Dust (CKD)	Cement kiln dusts (CKDs), including bypass dusts (CBDs) consist of alkali-rich particulate matter removed from cement kiln exhaust gasses			
4	Coal Combustion – Furnace Bottom Ash (FBA)	Grate residue from coal combustion			
5	Coal Combustion – Pulverised Fuel Ash (PFA)	Flue gas particulate matter from coal combustion			
6	Metallurgical (MTL)	Residues from metal production/processing			
7	Municipal Solid Waste Incineration (MSWI) – APC	Fine grained hazardous dust produced by flue gas treatment systems			
8	Municipal Solid Waste Incineration (MSWI) - Bottom Ash (BA)	Coarse grained residue remaining in the incineration zone after combustion			
9	Municipal Solid Waste Incineration (MSWI) - Fly Ash (FA)	Airborne particulate matter carried in the combustion gas and collected in the emissions filtration system			
10	Municipal Solid Waste Incineration (MSWI) - Boiler Ash (BLA)	Other residues arising from municipal incineration	3		
11	Paper Sludge Incineration Ash (PSIA)	Incineration residue from paper making wastewater sludge			
12	Sewage Sludge Ash (SSA)	Ash from incineration of wastewater treatment sludge			
13	Steel Slags (SS)	Metallic waste from steel manufacture			



Figure 1: Carbon dioxide uptake of wastes (dots denote means, whiskers illustrate minimum and maximum values)

Figure 2 illustrates the theoretical CO_2 capacity (determined using the Steinour formula), and the experimentally determined CO_2 capacity of the wastes. The disparity between the theoretical and experimental values should be noted. Thus, a waste that might be initially expected to be suitable for carbonation, based upon a preliminary assessment of its chemical properties, may in fact be non-reactive. As reflected in the variable chemical compositions, the CO_2 capacities both within and between the waste groups were also highly variable (see Figure 2).



Figure 2: Carbon dioxide uptake of wastes (dots denote means, whiskers illustrate minimum and maximum values)

The relationship between oxide composition and CO_2 capacity is being evaluated statistically to improve the prediction of CO_2 reactivity of wastes. Early results indicate that an equation model developed from the dataset is more accurate than previous models.

3 Product Development

Products were manufactured using a combined carbonation-induced forming technique described previously (Gunning *et al.*, 2009). Adjustment of the process parameters and mix formulations allows control over the grain size and shape of the carbonated product. Typical carbonated materials are shown in figure 3. Aggregate development was carried out in two phases. Firstly, aggregates were manufactured according to a standard set of formulations. These were subjected to basic testing (strength, leaching). The results were used to identify the aggregates conforming to the designed specification. Secondly, selected compliant aggregates were manufactured in larger quantities, and subjected to more rigorous testing.



Figure 3: (a) Typical raw waste, (b) Granulated carbonated product (c) Carbonated aggregate product

These selected aggregates were tested against the product specifications developed by Carbon8. Aggregates derived from municipal solid waste incineration air pollution control residues (MSWI-APCr), paper sludge incineration ash (PSIA), and biomass bottom/fly ash (BBA/BFA), were selected for further testing (EN 1097, EN 13055, EN 12390, ASTM C177-13). Alongside the three prototypes, a control chert gravel as an example of primary aggregate (1° Aggregate) and an expanded clay secondary aggregate (2° Aggregate) were also tested (see Table 3).

	Test	MSWI- APCr	PSIA	BBA/BFA	1° Aggregate	2° Aggregate
Aggregate	Apparent Density	2.6	2.5	2.5	2.7	2.0
	Bulk Density (kg/m ³)	1113	855	1050	1489	700
	Water Absorption (%)	16.1	24.1	17.8	< 0.5	19.9
	Crushing Resistance (N/mm ²)	8.3	5.2	6.4	20.3	6.6
	Thermal Conductivity (W/m.K)	0.2	0.15	•	0.15	0.24
Concrete	Bulk Density (kg/m ³)	1956	1702	1898	2121	1558
	Flexural Strength (N/mm ²)	2.7	2.9	2.8	2.9	3.7
	Compressive Strength (N/mm ²)	12.5	14.3	12.2	28.4	11.1
	Thermal Conductivity (W/m.K)	0.7	0.6	•	0.9	0.6
	Specific Heat (J/kg.K)	755	828	•	815	739
	Thermal Effusivity (J/m ² .K.s1/ ²)	1031	943	•	1285	792

Table 3. Aggregate/concrete testing results

• test results pending

The prototypes exhibited bulk densities akin to expanded clay, and were within BS EN 13055 requirements for lightweight aggregate. Crushing resistance was comparable to expanded clay, but considerably less than that of the chert gravel. The density of the concretes produced from the prototypes fell between that produced using expanded clay (1558 kg/m3) and from chert gravel (2121 kg/m3). Compressive strength of the prototype aggregate concretes was commensurate to expanded clay concrete, but exhibited lower flexural strength. The thermal properties of the prototype aggregate concrete was comparable to that of the expanded clay, thus was superior compared to the chert gravel concrete. These results indicate that the prototype aggregates have potential applications in the lightweight/medium-weight aggregate market.

A full end-use evaluation has been made for the MSWI-APCr aggregate, by validating its use in concrete construction block manufacture (see Table 4). The results show comparable properties between the standard product and MSWI-APCr aggregate substituted one in terms of strength (compressive and transverse failure), density, and geometry (flatness and parallelism). The MSWI-APCr aggregate has been granted end-of-waste status in the UK, and is now in full commercial production (currently 60,000 tonnes per year).

4 Pilot Scale Trials and Waste Declassification

Following a proof-of-concept trial planned to manufacture a bulk sample the BBA/BFA aggregate, an identical end-use evaluation for its use in concrete construction blocks will be undertaken. It is expected that the results of this work will be a major step forward in achieving end-of-waste status for this material.

5 Summary

The SAPICO2 project is developing new technology for combining gaseous CO_2 with solid waste to manufacture carbonate-cemented eco-construction materials:

- The project is a collaboration between the University of Greenwich (UK), the University of Picardie Jules Verne (France), and Carbon8 Systems (a spinout company from the University of Greenwich)
- SAPICO2 is a European Regional Development Fund project, supported by the Interreg's Manche (Channel) programme
- The project has sourced over 100 industrial wastes from across the UK and France
- The wastes have been chemically characterised and assessed for their carbon capture potential

- Wastes with high carbon capture potential have been developed into prototype products using accelerated carbonation technology
- A series of scaled-up trials have been conducted to produce bulk samples of selected prototypes and tested to international standards

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