

Construction Materials

The potential for using geopolymer concrete in the UK

--Manuscript Draft--

Manuscript Number:	
Full Title:	The potential for using geopolymer concrete in the UK
Article Type:	Limesnet Themed Issue
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Abstract:	Geopolymers are novel class of inorganic polymers which have potential to replace Portland cement in a number of different applications. Geopolymers can utilise a higher level of industrial by-products than Portland cement blends and numerous studies have concluded geopolymer concretes have significantly lower embodied CO2 than Portland cement based concretes. This paper examines the potential for the use of geopolymer binders as a Portland cement replacement in the UK. The quantities of material required, the major sources of these materials, the environmental implications and the barriers to implementation are discussed.
Additional Information:	
Question	Response
Please enter the number of words in your main text.	4799
Please enter the number of figures and tables in your submission.	5 Figures, 3 Tables

Introduction

Geopolymers are a novel class of inorganic binders and cements that can be used to form concrete and for other applications. They are formed by activating an aluminosilicate powder (precursor) with an alkaline solution (activator). The precursors can come from a range of aluminosilicate sources with various ratios of aluminium (Al) and silicon (Si) and include natural clays and other natural minerals, calcined clays such as metakaolin and industrial by-products such as fly ash and slags. Activators are typically sodium and potassium hydroxides (NaOH and KOH) and/or sodium silicate, but other alternatives are available. The term “geopolymer” is very broad and can be used to describe a range of products with a similar underlying chemistry. In the construction industry, the range of geopolymers can include materials used as an alternative to Portland cement (PC) based binders to those used as an adhesive with carbon fibres in strengthening applications.

This paper is focussed on geopolymers in high volume applications where they can be used as either a replacement binder in concrete or as an alternative to other high energy processes (for example as an alternative to firing in brick or tile manufacture). This paper will not describe the underlying chemistry and manufacture of geopolymers in detail as this is available elsewhere (e.g. Davidovits, 2011 and Duxson *et al.*, 2007).

The reasons for using geopolymers as an alternative to PC based binders are partly environmental and, depending on economies of scale, also cost reasons; however there are also significant technical merits to their use. Industrial by-products and waste materials which may or may not be suitable for use in PC based concretes can be used as geopolymer precursors, and a number of previous studies have shown that geopolymers can have considerably lower embodied carbon than PC based concretes.

Background

Most research and development was undertaken on a limited scale in Western Europe in the first half of the 20th century and later in the former Soviet Union where “soil cements” were undergoing research and use in the 1950’s. Research accelerated after the 1970’s when a French chemist, Joseph Davidovits, developed the term “geopolymer” and produced a large body of research into these materials.

Since that time, research has increased internationally with currently over 100 active research groups. The number of papers and books listed by Google Scholar as being published in a particular year is illustrated in Figure 1, showing the increase in paper titles containing “Geopolymer” and “Portland cement” since a baseline of 1980 where a value of 1 was assigned for each. In 2010, there were still considerably more papers on PC published, but the rate of increase for geopolymers is considerably higher with an increase of 100 times in the 30 year period compared with an increase of 6.6 times for Portland cement. While the analysis may not be exact, as some papers on the subject could be published without those terms in the title, it does demonstrate the relative increase in research in the two areas.

Depending on the precursors and mix design, geopolymer concrete can appear similar to PC-based concrete in both fresh and hardened states, but the underlying chemistry between the two is fundamentally different. Geopolymers are composed of an amorphous three-

dimensional network comprising mainly aluminium (Al) and silicon (Si) oxides, while PC has a semi-crystalline gel binding phase (Lecomte *et al.*, 2012), with higher calcium content. The terminology used can be varied and “geopolymers” could more generally be described as inorganic polymers or alkali activated materials (AAMs). There is some conflict over the terminology as some researchers have indicated that the term “alkali” brings connotations of alkali-silica reaction (ASR) to those with a background in PC-based concrete, even though geopolymers are generally resistant to ASR during accelerated tests such as RILEM AAR-3. This paper will focus on materials with a low calcium content and uses the framework in Figure 2 which is similar to that to be included in the report by RILEM Technical Committee 224-AAM on Alkali Activated Materials.

Current material use

In order to assess the potential for geopolymers in bulk construction applications, it is necessary to quantify the market of existing similar products. The data shown in Figure 3 were obtained from BIS (2012) and include UK material production figures for cement, concrete block (combined dense, lightweight and aerated) and clay brick. In addition, the relative material price increase from 2005 is indicated, based on a value of 100 for 2005 for each of the materials. The material quantities in the figure were calculated based on a series of representative densities of the concrete blocks of 1800, 1300 and 700 kg/m³ for the dense, lightweight and aerated blocks and based on a fired brick unit mass of 2.35 kg, as the quantities are provided in inconsistent units in the original reference (for example, the number of bricks, rather than total mass, is provided).

The economic downturn which started in 2007 had a significant impact on the quantity of materials produced, with clay brick production reacting to the downturn before cement and concrete block production. Some of the cement produced would have been used in the production of the concrete blocks shown in Figure 3, but the majority of the cement produced would have been used for other applications including ready-mix concrete and precast concrete component production. Based on the data in Figure 3, the reduction in demand did not create a corresponding reduction in cost with prices generally increasing above the rate of inflation. Using the Composite Price Index from the Bank of England, the relative cost would have increased from 100 to 119 between 2006 and 2011.

Potential precursor sources

If geopolymers can be manufactured with a much lower carbon footprint than PC, in order to have a significant impact on UK carbon emissions, it would be necessary to replace significant quantities of PC with geopolymer binder. For this to occur a number of technical, capacity and economic requirements have to be met, the first of which is that there needs to be sufficient material available for use as geopolymer precursors.

The EU has developed a list of 14 different minerals which are considered economically important but are subject to a higher risk of interruption of supply, and the use of these minerals is considered inappropriate for use in geopolymers or other bulk material applications. These minerals are listed in alphabetical order in Table 1 (EC, 2010).

With the critical minerals excluded from potential precursors for use in geopolymers, realistic alternatives could be either geologically based (natural) minerals, or waste materials. Only the major potential sources are included here.

Geologically based minerals

Much of the original work on geopolymers by Davidovits focussed on using metakaolin, which is relatively pure kaolin (also called china clay) that has been “calcined” or heated until dehydroxylation. Kaolin is a 1:1 clay mineral with one layer of silicate and one layer of alumina and, unlike the limestone used in PC manufacture, there are no significant quantities of calcium present. For applications in geopolymers or as a pozzolan in PC based mixes, the kaolin is normally heated to approximately 750°C and then ground to form metakaolin. This heating and grinding does increase the embodied energy and embodied CO₂ of the material, but as there is no carbon present in the kaolin, carbon dioxide is not directly emitted as occurs when lime or cement are manufactured. Geopolymers based on metakaolin can achieve the same strengths as commercially available PC concretes but the fresh properties may differ.

The UK was the sixth largest global kaolin producer in 2009, with almost all extraction in the Cornwall/Devon area. Modern kaolin extraction is subject to stringent environmental controls and management plans which limit the environmental impact of these activities, but the landscape in parts of the south west of England shows considerable evidence of historical extraction. UK kaolin extraction reduced considerably between 2005 and 2009 (from 1.9 to 1.1 million tonnes) while in the fifth largest global producer in 2009, Iran, production increased from 0.5 to 1.2 million tonnes over the same period (BGS, 2011). The reduction in UK extraction over this period was not because of shortage of supply but rather a result of cost effectiveness. The size of reserves is not published because of the commercial nature of this information, but more than 50 year’s capacity is available using current technology (BGS, 2011). UK china clay production peaked in 1988 at nearly 2.8 million tonnes, but only a small portion of this would have been processed to metakaolin, with the major uses of china clay as paper fillers and in the ceramics industry. Based on the difference between the peak and current capacity, it is estimated that at least an additional 1.6 million tonnes of dry kaolin could be sourced per year for geopolymer production, which because of dehydroxylation, would amount to approximately 1.4 million tonnes of metakaolin.

In addition to the relatively pure kaolin described above, the UK possesses considerable kaolin reserves used for brick manufacture. UK brick production has been in decline since the 1970s with almost 16,000 million tonnes of brick clay extracted in 1974 (BGS, 2007), decreasing to around 4,000 million tonnes in 2011, as shown in Figure 3. What the brick industry describes as brick clay is not geologically clay, but mainly silt and sand with approximately 30% clay sized particles (Heath *et al.*, 2009). The clay minerals preferred in brick manufacture are kaolinite and illite (BGS, 2007) and both have been successfully used for geopolymer production. Taking the 12,000 million tonne difference in extraction between 1974 and 2011, estimating the clay content of this as 30% of the total mass, there should be at least 3,600 million tonnes of mainly kaolin and illite available per annum from current and closed brick extraction pits. Geopolymers using uncalcined kaolin or illite are typically a lower strength material and may be more suited for use as a brick or concrete block alternative, rather than as structural concrete (Davidovits, 2011).

Based on the 12,000 million tonne difference between 1974 and 2011 annual extraction, there is potential to replace all the approximately 8,000 tonnes of concrete blocks used in the UK. There is also scope for brick clay to be calcined and ground into an impure form of metakaolin / calcined illite, but this may increase energy beyond that required for PC manufacture.

In addition to the kaolin extracted as almost pure “china clay” and that used in brick manufacture, there are a number of other sources of kaolin and other suitable clays, including from “ball clay” extracted in the Devon area, and extraction from other weathered materials and quarry or mining wastes throughout the UK.

Industrial by-products

Recent research into geopolymers has focussed on the use of industrial by-products, most notably fly ash and various slags. There are numerous other aluminosilicate by-products that could be used for geopolymerisation, but fly ash and slags have the benefit of extensive heating during processing which can promote an amorphous (more reactive) structure which is utilised when used as an additive to PC blends and when forming geopolymers.

A number of different slags are produced in the UK and some of these may be suitable for geopolymerisation. The main slags currently produced are blast furnace slag and steel slag, although smaller quantities and historic stockpiles of other slags are present. Disadvantages of using historic stockpiles include the variability in the properties and the potential change in properties as a result of leaching and carbonation since the slags were originally produced. The majority of research into slag use for geopolymers is focussed on ground granulated blast-furnace slag (GGBS). As the calcium content in GGBS is higher than for other common precursors, it may fall outside the “geopolymer” block in Figure 2, but as an alkali activated material it is still included in this paper. Approximately two million tonnes of GGBS are produced each year in the UK (CSMA, 2012), and almost all of this is used as an addition in blended PC based concretes. This leaves little available for geopolymer cement production unless geopolymer concrete replaces some PC based concrete.

Approximately six million tonnes of fly ash or pulverised fuel ash is currently produced in the UK each year, of which approximately 50% is sent to landfill (WRAP, 2008). In addition, there are significant stockpiles of fly ash of variable quality around the UK, with approximately 114 million tonnes accessible in stockpiles. Fly ash has been extensively used as a precursor in geopolymer production and this is probably the most significant current UK resource for geopolymer production. The largest current use of fly ash is in the manufacture of construction products based on PC.

The use of fly ash in PC based concrete is governed by the European standard EN 450: “Fly ash for concrete”, and this includes specifications for fly ash from co-combustion of coal and biomass, with one limit of a minimum 80% coal by dry weight in the fuel mix. This presents a particular challenge in the UK context as government predictions indicate a significant decrease in coal based electricity supply and a corresponding increase in renewable supply, as shown in Figure 4 (based on DECC, 2011). The figure combines conventional and carbon capture and storage (CCS) for both coal and gas based electricity, and includes electricity from combined heat and power (CHP) plants. The renewable category includes all forms of renewable energy, including biomass, solar and wind, and it is predicted that biomass will

account for 25-45% of the renewable generation by 2020. The use of biomass ash in geopolymers shows particular promise as biomass ash is generally higher in potassium than coal ash, potentially reducing the amount of activator required.

There is also potential for other smaller waste and by-product streams to be used, including paper sludge ash which is particularly rich in amorphous aluminosilicates from the combustion of the wood fibres, but also from the dehydroxilation and calcination of the paper fillers (particularly kaolin/china clay) during combustion.

The current sources of major precursors are summarised in Table 2. These are based on current production, even though production of coal-based fly ash is predicted to decrease while production of biomass based fly ash is expected to increase.

If Figure 3 and Table 2 are viewed together, it can be concluded that there is potentially sufficient material to replace the approximately 10 million tonnes / year of current UK cement use with metakaolin, fly ash or GGBS based geopolymer cement if some existing fly-ash stockpiles are used. As UK coal-based fly ash production is expected to decrease, there may not be sufficient PFA to use as precursors in the future, unless the increasing use of biomass ash is utilised or PFA is imported into the UK.

There are, however, a number of reasons why a complete replacement of PC mixes is unlikely, and these include the versatility and ease of use of PC compared with current formulations of geopolymer cements. Similarly, although there are sufficient precursors available to replace all fired brick and concrete blocks currently used, it is unlikely this will occur as the aesthetic characteristics of fired brick cannot be easily replicated with geopolymers, even if the same source materials are used.

Environmental considerations

Environmental considerations include a number of different factors such as use of waste materials, primary resource use, embodied energy, embodied CO₂ and environmental toxicity. This section is focussed on use of waste materials and embodied carbon.

Use of waste materials

As mentioned earlier, almost all the GGBS and approximately half the fly ash produced in the UK is currently used, with the other major precursors in Table 2 from geological sources and therefore not from waste streams. If fly ash use remains constant and coal based electricity supply follows the UK government predictions in Figure 4, it is likely that all new fly ash produced in the UK will be used from approximately 2022, and additional fly ash will have to be extracted from stockpiles as is currently underway in some other European countries. If fly ash is used as a precursor in geopolymers, it will be used in higher percentages than in blended PC concrete, resulting in stockpiles having to be utilised earlier.

Any pure biomass ash or ash from biomass co-fired with coal at ratios unacceptable to EN 450 will have to be either disposed of or a beneficial use for it found. Geopolymers based on fly ash which does not meet the requirements of EN 450 or on PSA may be one beneficial use of these materials, but this will require further investigation

Whether a particular material is considered a waste is relevant to its calculated emissions, as co-products can be allocated a portion of embodied energy and carbon, while wastes are generally assigned no emissions. The assumptions of allocation of embodied CO₂ are becoming increasingly important with the 2008 EU Directive 2008/98/EC which states that a material may be classified as a by-product rather than a waste if “(a) further use of the substance or object is certain; (b) the substance or object can be used directly without any further processing other than normal industrial practice; (c) the substance or object is produced as an integral part of a production process; and (d) further use is lawful”.

Embodied carbon

The UK has committed to reducing carbon emissions by at least 80% below 1990 levels by 2050. Concrete production accounts for approximately 7% of global CO₂ emissions (Basheer, 2009) which is considerably greater than that from aviation (Meyer and Tol, 2010). While this figure is lower for the UK because the UK is a net PC importer, the change from the almost universal use of PC based concrete to a concrete with significantly lower embodied CO₂ would have a marked effect on UK emissions. The extent of this reduction would depend on the availability, uptake and embodied CO₂ of the replacement concrete.

Life cycle assessment (LCA) methodology and details are beyond the scope of this paper, and all information presented here is based on simplified cradle to factory gate analyses as there is insufficient information on the full lifecycle, including end-of-use, of geopolymer concrete or other geopolymer building products.

Rather than performing a full analysis of the embodied carbon of geopolymers, the percentage reduction in embodied carbon for either the binder only or concrete from recent sources in the literature is summarised in Table 3. These references do not specifically include heating during curing as many modern geopolymer mixes do not require heat curing, which is often impractical for on-site applications. As far as the authors are aware, no study has investigated the environmental implications of a reduction in activator coupled with an increase in heating during geopolymer curing.

As shown, all references show geopolymer concretes have lower emissions than PC-based alternatives and all references showed the activators have the largest contribution to embodied CO₂. Since the embodied CO₂ data for sodium silicate used in all references was developed in 1995 for the European situation (Fawer *et al.*, 1999), average CO₂ emissions per unit electrical and heat energy have reduced by over 20% in Europe (IEA, 2011), which implies the benefit of using geopolymers may be even greater than indicated.

The reference with the lowest reduction (Habert *et al.*, 2011) assumes a neutral sodium silicate (SiO₂:N₂O weight ratio of 3.3) activator instead of an alkali sodium silicate activator (SiO₂:N₂O ratio of 2) which is more beneficial for geopolymerisation. This assumption artificially increases the sodium silicate activator embodied CO₂ emissions by 48%, based on the data included in the LCA database used for the paper (Fawer *et al.*, 1999). As the sodium silicate contributed approximately 70% of the embodied carbon in the Habert *et al.* analysis (for fly ash considered as a waste) and with an error of almost 50% in this value, the calculated emissions in this paper are considered erroneously high, and are therefore not used in the subsequent analysis.

Based on the data in Table 3, and with PC-based concrete production accounting for approximately 7% of global CO₂ emissions, the likely range of global CO₂ reductions can be calculated based on the percentage PC concrete replacement with geopolymer concrete. The range is calculated using the maximum and minimum values from Table 3 (excluding the reference by Habert *et al.*, 2011), all of which include no allocation of emissions for precursors. This is a correct assumption for this particular analysis as the intention is to show the potential for change in actual CO₂ emissions and allocation of emissions has no effect on actual emissions.

Although it is unlikely that there will be a 50% replacement of PC based concrete with geopolymer concrete in the near future, the effect would be considerable and could approach the total CO₂ emissions of all global aviation which is currently approximately 3% of global emissions (Meyer and Tol, 2010).

The embodied carbon of geopolymers could be further decreased if lower embodied carbon sources of activators are found, which could include use of by-products in activators and lower carbon forms of electricity, the main component of the embodied CO₂ in sodium silicate activators (Fawer *et al.*, 1999). As carbon dioxide is directly produced during the manufacture of Portland cement (approximately 0.55 tonnes CO₂ chemically emitted per tonne PC produced, with the remainder processing emissions), there is little scope for these same large reductions in embodied CO₂ for PC.

Barriers to geopolymer use

At the time of writing, there is only one company in the UK extensively marketing geopolymer building products, but the market is more developed elsewhere, particularly in Australia. The UK construction industry is notoriously conservative and this may be a significant barrier to geopolymer uptake. The 1998 Egan report on the UK construction industry (Egan, 1998) stated: "There is already frustration amongst component suppliers that their innovations are blocked because construction workers cannot cope with the new technologies that they are making available." If geopolymers are formulated to be used in the same manner as conventional concretes, either through mix design or use of precast elements, this will aid acceptance by construction workers, but designers are reluctant to specify materials where no standards exist.

This is a point of particular concern for geopolymer concretes where no nationally adopted standards currently exist, although RILEM committee 224 recently completed work on standardising performance testing procedures for these materials. Performance standards are not always compatible with EU approaches where standard EN 206-1:2000 for "Specification, performance, production and conformity of concrete" states that concrete must gain strength by hydration of cement on mixing with water. This limits the use of alternative cements, even if they demonstrate considerable environmental and/or technical benefit.

Education is a further barrier to geopolymer cement uptake as these materials are not currently at a "just add water" level of technology. It is unrealistic to assume undergraduate civil engineering students in the UK have the inorganic chemistry background required to understand the geopolymeric reactions and, more importantly, the implications for

carbonation and other aspects of durability. It is unlikely that the difference between PC and geopolymer concretes is currently covered in detail in any undergraduate civil engineering programme in the UK.

Conclusions

Geopolymers have the potential to significantly decrease global CO₂ emissions. Reductions of this magnitude are not available to PC manufacture because of the chemical release of CO₂ during heating of limestone during Portland cement production. Geopolymers can be based on abundant natural and industrial waste materials (mainly GGBS and fly ash) which can increase environmental benefits. Because of the ease of use, it is unlikely that Portland cement will cease to be used in the UK, but there is potential for geopolymer binders to replace a significant portion of current Portland cement use.

Initial routes to market for geopolymer concrete products in the UK could be through precast elements (including blocks), as the risks are lower and control of production and curing higher than for in-situ mixes. Education on geopolymers and other low-carbon concrete alternatives should be included in university programmes for both undergraduate and postgraduate courses, and for those working in the construction industry.

There may not be sufficient PFA meeting EN 450 and GGBS produced in the UK if geopolymer cement were to replace a significant portion of the current Portland cement production, largely because coal power generation is expected to significantly decrease in the future. It is likely that existing stockpiles of PFA will have to be mined to meet blended Portland cement requirements in approximately 10 years, and even before then if geopolymer concretes based on fly ash are used extensively. Almost all GGBS currently produced in the UK is already used, the majority of this in blended Portland cement mixes. The options are therefore to either continue to use PC-based concretes with a relatively low fly ash / GGBS content (and with high CO₂ emissions), import fly ash and GGBS into the UK for geopolymer production, or investigate other suitable materials for use as geopolymer precursors.

Other potential precursors could include new or existing waste streams with suitable amorphous aluminosilicates, or geologic alternatives. Potential new waste streams include co-fired or biomass fly ash and paper sludge ash, but the use of these new waste streams in geopolymers would require extensive research. Geologic alternatives should be investigated for the UK to ensure long-term security of supply.

Acknowledgements

This paper is based on a study mission funded by the UK Engineering and Physical Sciences Research Council (EPSRC) through the Low Impact Materials and innovative Engineering Solutions research Network (LimesNet). LimesNet is an international multi-disciplinary community of leading academic researchers, industry members and other stakeholders, who share a common view for the development and adoption of innovative low

impact materials and solutions to deliver a more sustainable built environment in the 21st Century.

The authors would like to thank Joseph Davidovits from the Geopolymer Institute and Yiannis Pontikes and colleagues from the Katholic University of Leuven for their valuable time and insights into geopolymer technology and applications. Information provided by John Provis from the University of Sheffield is also gratefully acknowledged.

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Table 1: List of critical raw materials or mineral groups at EU level (EC, 2010)

Antimony	Indium
Beryllium	Magnesium ¹
Cobalt	Niobium
Fluorspar	PGMs (Platinum Group Metals)
Gallium	Rare earths
Germanium	Tantalum
Graphite	Tungsten

¹ Magnesium is regarded as critical, but magnesite (magnesium carbonate) was investigated and not included on the list of critical minerals.

Table 2: Summary of major potential precursors for geopolymers in the UK, based on current production

Potential precursor	Quantity available (million tonnes/year)*	Potential geopolymer products
China clay / metakaolin ¹	At least 1.6	Cement for concrete / blocks
Brick clay	At least 3.6	Geopolymer blocks
GGBS ²	2	Cement for concrete / blocks
PFA ³	6+stockpiles	Cement for concrete / blocks
<p>* References included in text. ¹ Based on difference between peak and current production. ² Almost all current production of 2 million tonnes/year used in PC based mixes. ³ Coal based PFA is predicted to decrease significantly over the next 10-20 years, but biomass ash likely to significantly increase.</p>		

Table 3: Geopolymer emissions as a percentage of PC based concretes or pastes

Reference	Location	Percentage embodied carbon compared with PC binder or concrete
McLellan et al, 2011	Australia	44-64
Habert et al, 2011 ¹	France	55-82 (higher figure for economic allocation of emissions from fly ash)
Davidovits, 2011	France	19 (no emissions allocation for GGBS)
Van Deventer et al, 2010	Australia	20-40 (lower figure is for binder only)

¹ Only fly ash geopolymers included, although GGBS and metakaolin also investigated

Figure 1

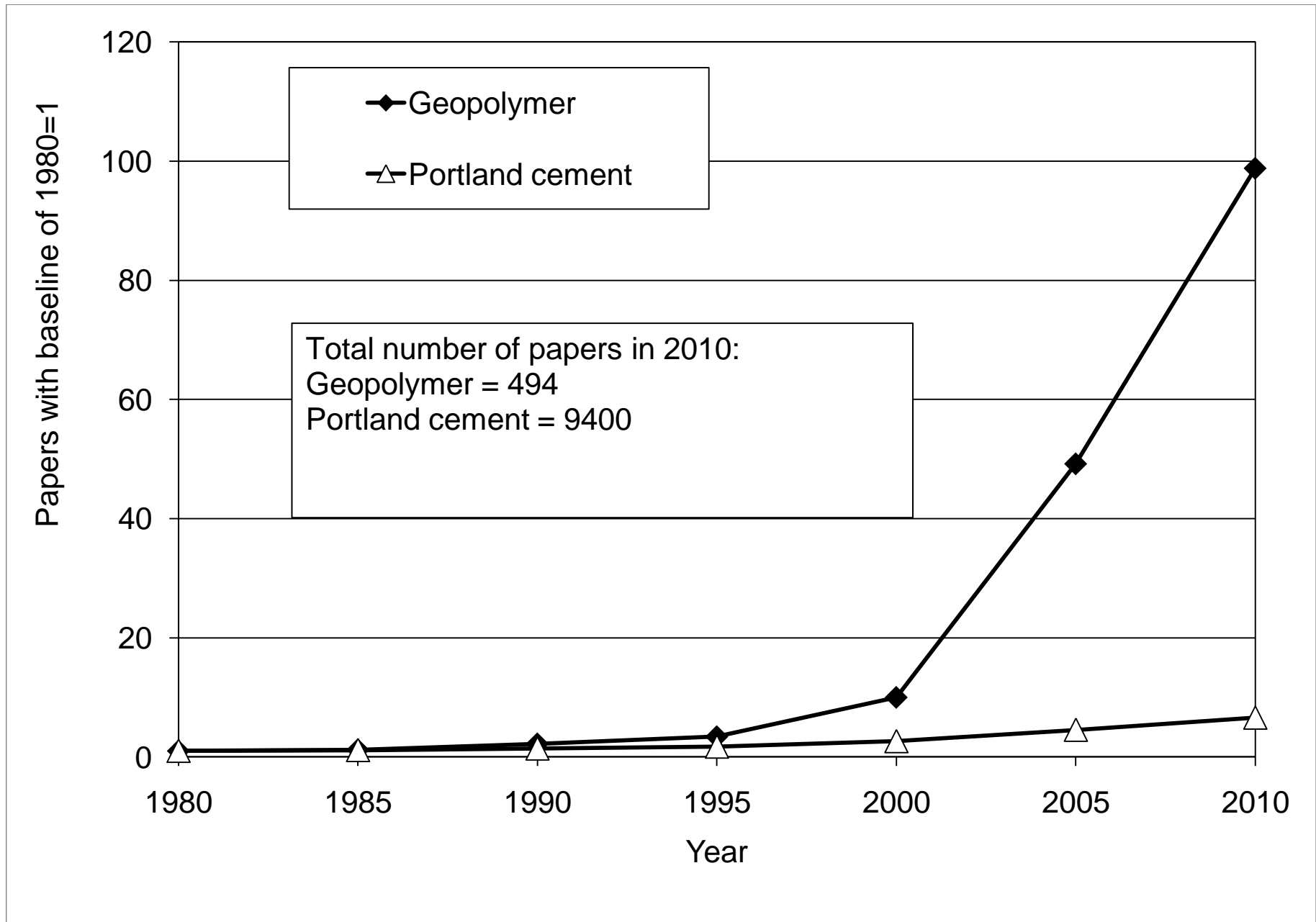


Figure 2

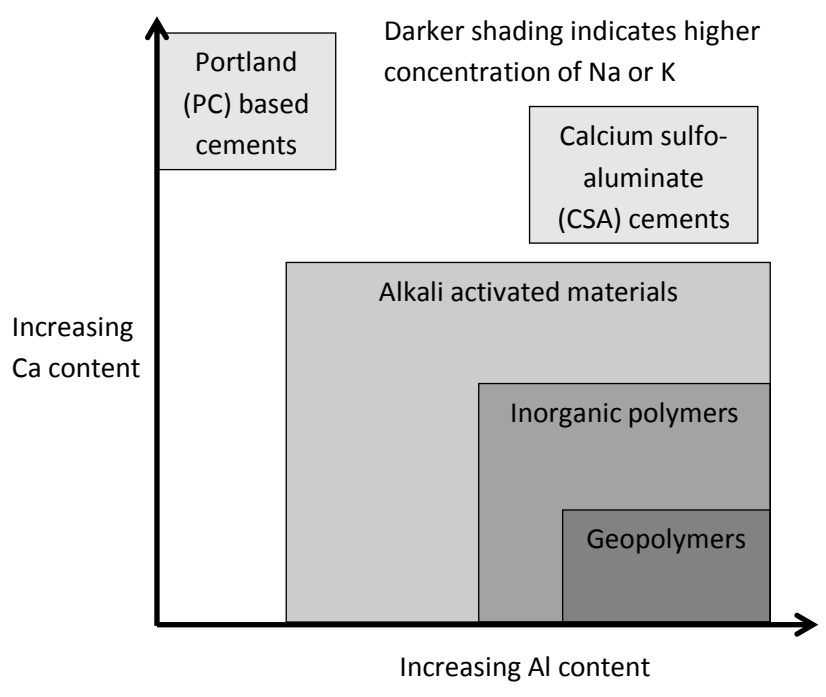


Figure 3

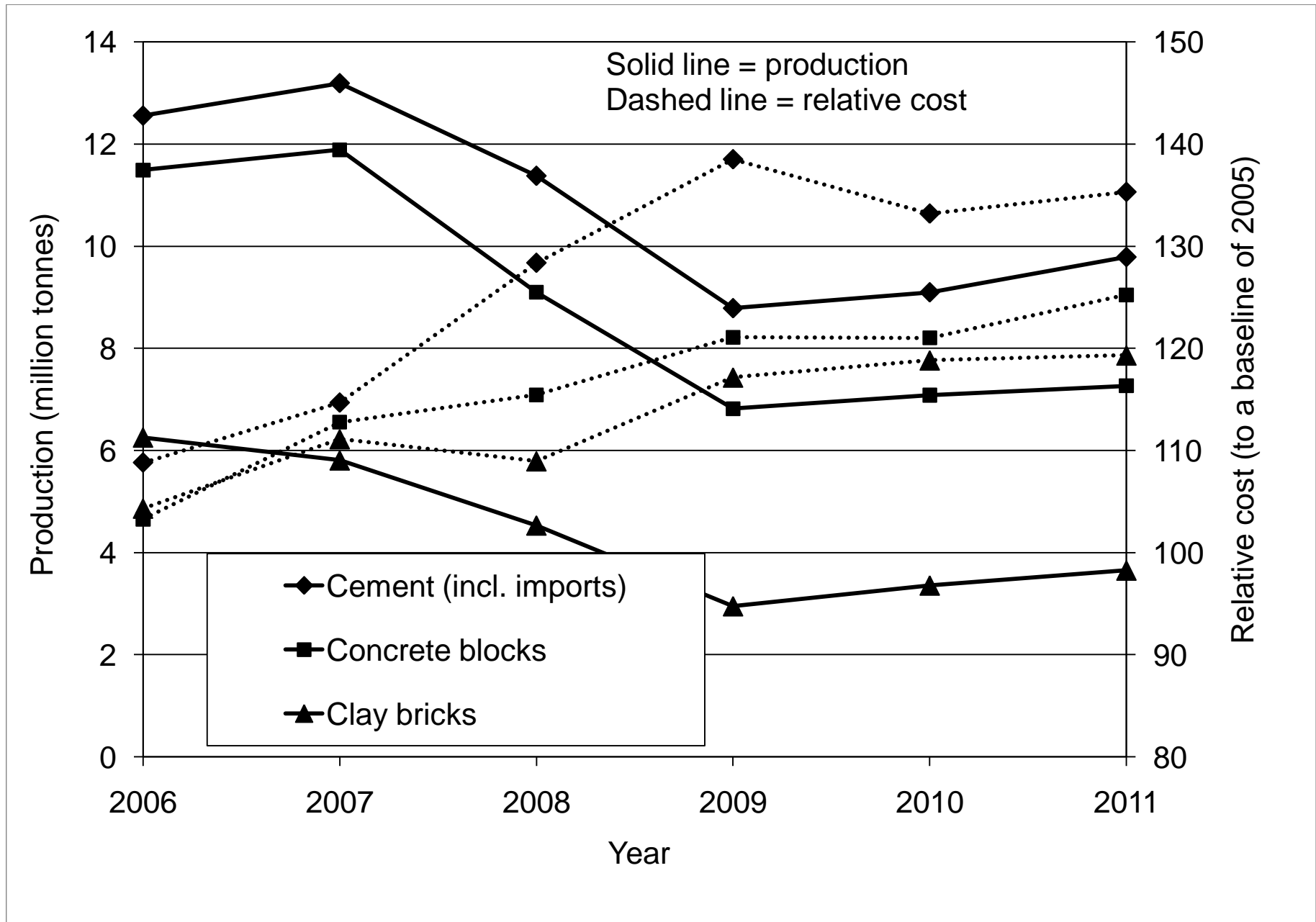


Figure 4

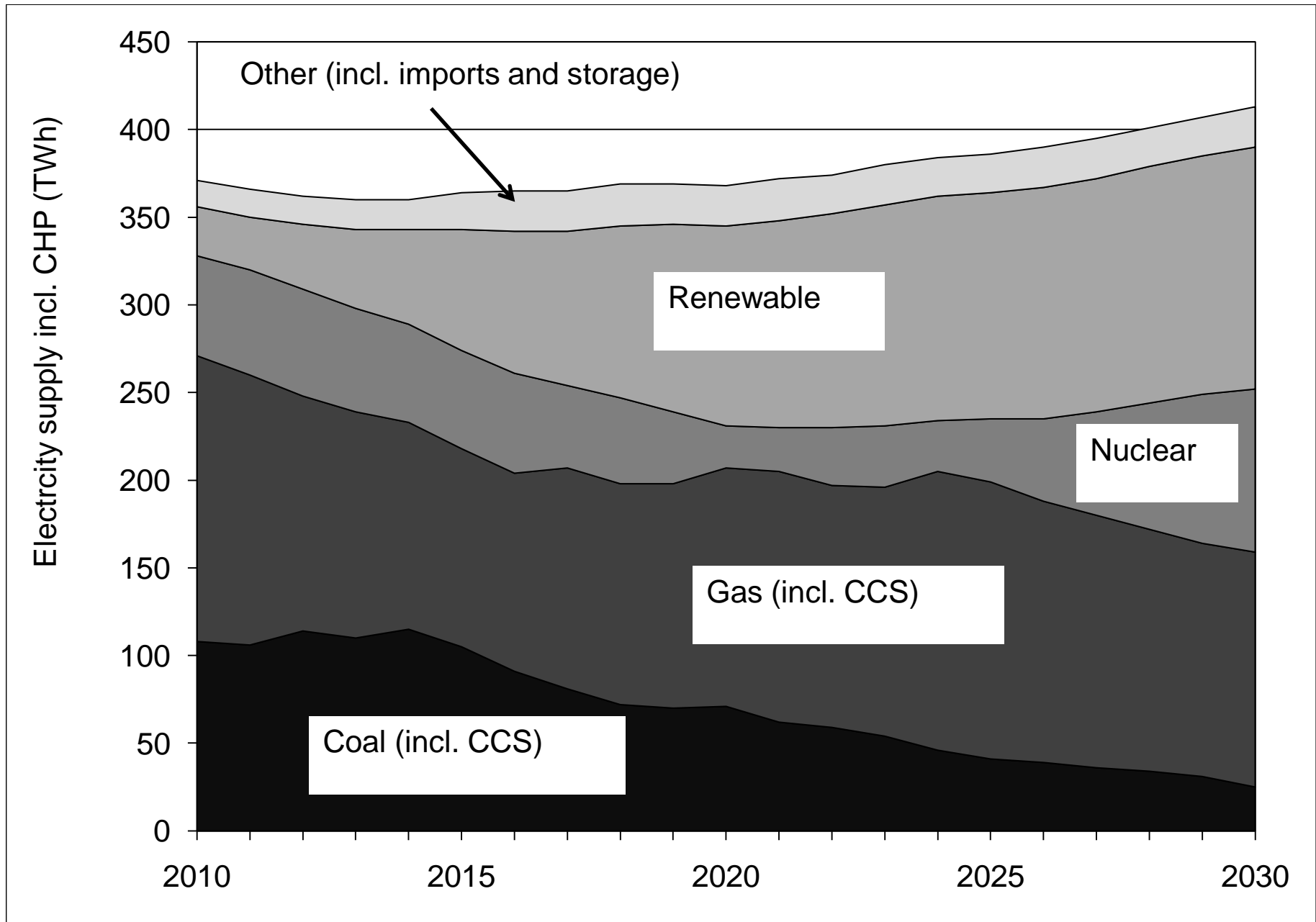


Figure 5

