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A brief critical view on the carbon-conditioning of recycled aggregate using pressure chamber

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Abstract

CO₂ Concrete as an environmental material had attracted the public view for many years. Several methods had been proposed for entrapping CO₂ gas in concrete some of them targeting cement paste as the reactant with the CO₂ gas and the other methods consider aggregate as the reactant. The common purpose of all, is to, first, reduce the impact of greenhouse gas emissions via the means of CO₂ gas consumption, and second, to increase the mechanical properties of concrete by accelerating the formation of calcium carbonate crystals. Seeking to achieve a successful wide scale commercialization, it is important to address the flawed aspects of a concept. This paper projects a critical point of view on some basic flawed assumptions taken as facts, in the conceptualization of carbon-conditioning of recycled aggregate.

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Keywords

CO₂ Concrete; Chemical absorption; Recycled aggregate

Introduction

With an increasing attention to the environmental impact of construction, scientist and the broad community of construction practitioners are making attempts to reduce the level of greenhouse gas emissions. Concrete as the most widely used material after water globally accounts for the largest portion of the CO₂ emission. Two main ingredients of concrete, being cement and aggregate, have the highest level of human exploitation of natural resources. And in order to reduce these harmful effects, the use of supplementary cementitious materials and recycled aggregate had become popular. Nevertheless, the environmental impact of this human made rock, concrete, is still considerably high. Governments across the globe are investing significantly on the new environmental technologies to lower the environmental impact of construction even further down. One of the emerging technologies in this area is called CO₂ Concrete.

Carbonation techniques

There are three major CO₂ concrete technologies proposed so far, each with their own unique procedures. Kou et al. [1] uses a vacuum/pressure chamber for the carbonation of recycled aggregate. In the procedure introduced by Kou et al., the chamber is vacuumed to -50 KPa before the CO₂ injection. The CO₂ pressure in the chamber is controlled by a regulator at 10 KPa with different carbonation period. In order to avoid the exposure of aggregates to humidity (which is claimed to be disruptive to the carbonation reaction) a silica gel is placed at the bottom of the chamber underneath the aggregates. The schematic view of this methods is shown in figure 1 below.

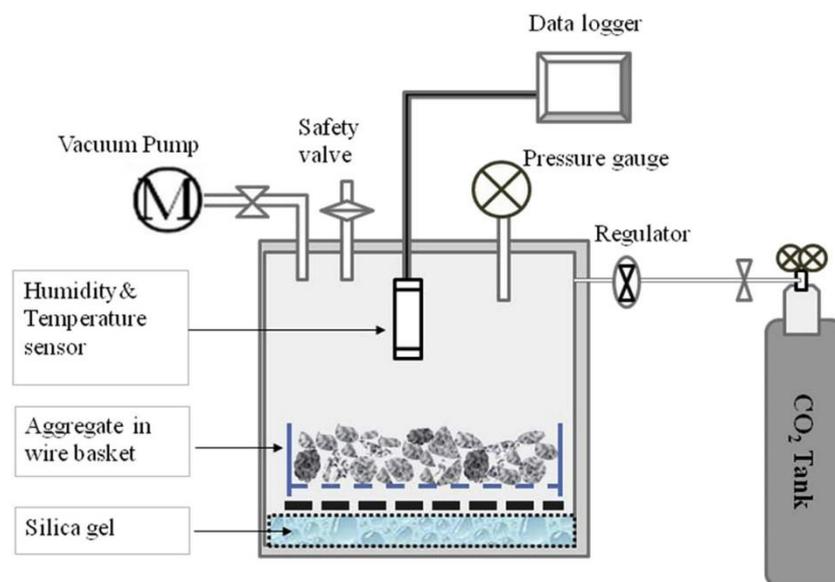


Figure 1 Schematic of CO₂ curing experiment by Kou et al. [1]

In another method proposed by Bao Jian et al. [2] hardened concrete blocks are placed in a tube where the CO₂ flows through. The tube has a diameter of 15 cm and a length of 40 cm. The flow rate is controlled by a regulator and ranges from 0.2-10 L/min depending on the experimental setting. The gas

outlet is connected to a limewater container to absorb the surplus CO₂. The figure 2 below illustrates the procedure in this method.

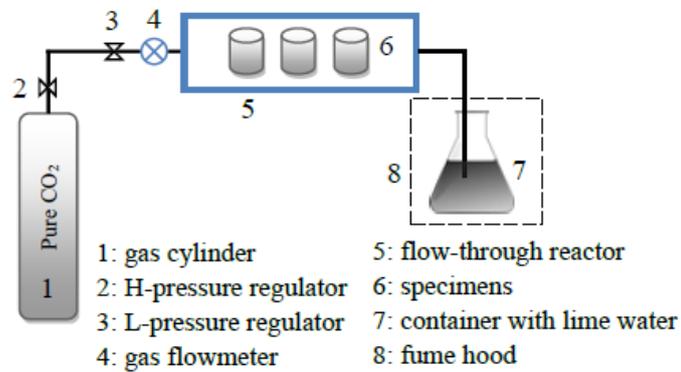


Figure 2 Flow-through CO₂ curing method proposed by Bao Jian et al. [2]

Butera et al. [3] introduces the similar procedure to Kou et al. except in his procedure, aggregates are exposed to a higher CO₂ pressure. Vacuuming the chamber and the presence of silica gel is also another major difference between these two methods. Figure 3 below shows the procedure introduced by Butera et al.

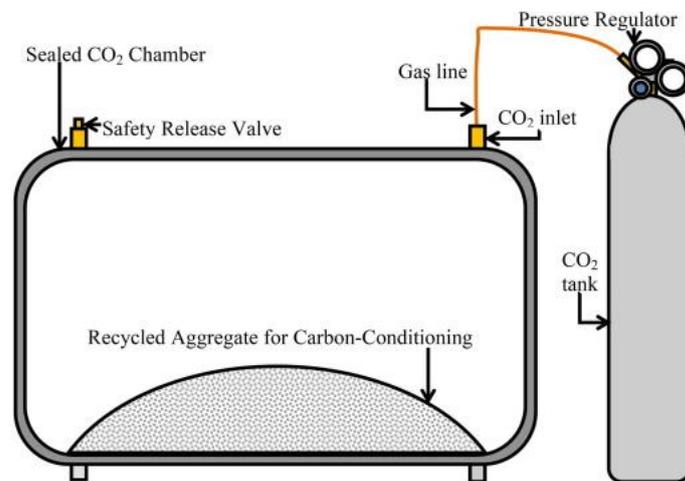
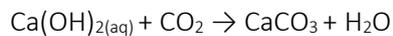


Figure 3 Schematics of carbon conditioning method proposed by Butera et al. [4]

Carbonation reaction

Carbonation process is a complex process of mass transfer and chemical reactions. Commonwealth Science and Industrial Research Organization (CSIRO) outlines that the CO₂ mineral carbonation is comprised of three distinctive steps respectively: leaching of solid matrix; CO₂ dissolution into solution; carbonate precipitation. The reaction demands stirring as the efficiency requirement [5]. The chemical reaction is as followed:



This reaction is zeroth order with respect to carbon dioxide concentration at ambient temperature [6]. A zero-order reaction is a chemical reaction wherein the rate does not vary with the increase or decrease in the concentration of the reactants. The rate of these reactions is always equal to the rate constant of the specific reaction.

The kinetics of the reaction requires an activation energy of 145 kJ/mol. Furthermore, this reaction occurs at the temperature of above 500°C [6-10]. CO₂ bond dissociation energy is reportedly given as +532.2 kJ/mol for a single oxygen and carbon bond in a CO₂ molecule [11].

Critical view

From the chemistry

Referring to the kinetics of the carbonation reaction, none of the CO₂ Concrete technologies mentioned before satisfies the conditions required for the reaction to occur. The so-called carbon-conditioning proposed by Butera et al. occurred at the ambient temperature with the varying pressure. Pressure variance in gaseous materials is equivalent to the change in the concentration of the CO₂. Whereas, carbonation is intrinsically a zeroth order reaction at the ambient temperature i.e. change of pressure does not influence the rate of reaction at ambient temperature. Furthermore, CO₂ covalent bond energy is fairly high. This molecule has a stable structure. At the exposure of this gas to the crystalline rocks the possibility of reaction is close to zero.

A negligible 3-10% increase in the compressive strength was reported in the literature and correlated with the formation of calcium carbonate crystals as a result of carbon-conditioning recycled aggregate [3]. Considering the schematics of figure 3 and the presence of moisture and pressure in the chamber, it is highly possible that formation of carbonic acid causes an slight acid treatment of recycled aggregate which enhances the properties of ITZ zone, unless human errors are neglected.

Petrography, scanning, and X-ray diffraction

This section is based on the (wrong) assumption of the occurrence of the carbonation reaction.

A recently published paper by Tam et al. [12] states the petrographic analysis shows that aggregates are completely carbonated. A complete carbonation means that all reactants of the carbonation reaction above are consumed and the product is a white substance known as calcium carbonate. Whereas, in reality, there has been no changes in color observed after the so-called carbonation of recycled aggregate. Even in an ideal condition for the formation of calcium carbonates, the growth of solid product islands occurs on the surface of grain or pore [13] and not in the core of the aggregate which is emphasized in the pictures reported in the same article. The reported pictures in the article shows a bright yellow colour even at the depth of the aggregate that are apparently digitally modified

(figure 4). It is more likely that the yellow colour intensity of the pictures had been modified by a software as in one plot cement reflects the yellow spectrum after the colour adjustment and in another plot, cement reflects a darker spectrum after the yellow colour intensity had been dimmed in the software.

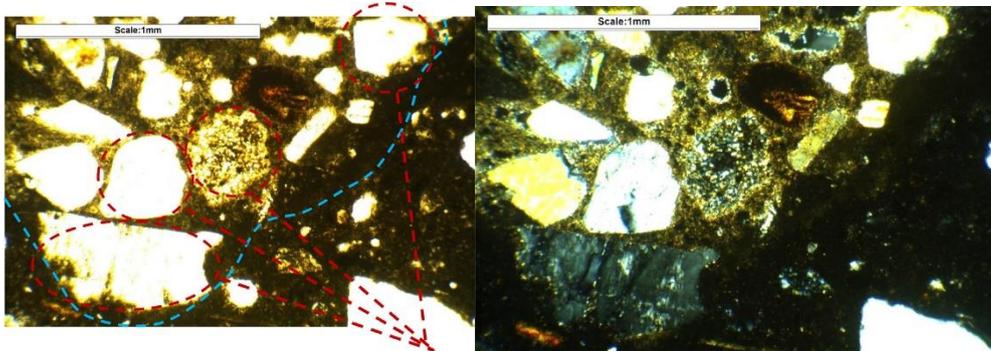


Figure 4 Petrographic results of so-called carbon-conditioning

There are two popular models for a general gas-solid reaction namely shrinking-core model and shrinking-pore model. The idea in both of these models is that the reactant material will be covered deep in the solid products as the reaction proceeds [14, 15].

The same study, in the results of x-ray diffraction indicates that crushed cement mortar has peak calcium carbonate content of 4 times higher than recycled aggregate which is not a new finding. Hydrated cement has a high portion of calcium carbonate regardless of the mixing method. Thus, the comparison is invalid.

Conclusion

Considering the kinetics of carbonation reaction which results in the formation of calcium carbonate crystals, the formerly proposed methods of carbon conditioning of recycled aggregate are invalid. There are major assumptions that are neglected in the conceptualization of the so-called CO₂ concrete using pressure chamber. First, the activation temperature for solid-gas reaction of calcium hydroxide and CO₂ is above 500°C. Cement kiln calcination, for example, occurs at the temperature of 900°C. Second, the microstructure analysis presented in one of the articles does not imply the shrinking-pore model of solid-gas reaction. Third, carbonation is intrinsically a zeroth order reaction which means that the rate of reaction is independent from the concentration of the reactants i.e. change in the pressure inside the chamber does not have any effect on the growth of calcium carbonate.

Overall, it is not a scientific statement to label the corresponding phenomena as carbonation reaction. Carbonation reaction does not occur in the proposed method. The most possible reaction is the formation of carbonic acid at the exposure of moisture to carbon dioxide which can slightly accelerate the acid treatment.

Conflict of interest

The author declares that, to the best of knowledge, there is no potential for a future conflict as all participants are keen on upgrading the current CO₂ concrete technology.

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