



Urea additives for reduction of hydration heat in cement composites



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HIGHLIGHTS

- Dissolving urea in water reduces the solution temperature abruptly and drastically.
- Mixing urea in cement composites can reduce the hydration-generated heat.
- The effects of urea mixing on structure and strength were assessed experimentally.
- The compressive strength and pore features of conventional mixes can be preserved.

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ABSTRACT

This study evaluates the feasibility of using urea in cement composites, as a means to reduce the heat generated from the hydration of cement. The performed experimental evaluation uses several different test and analysis techniques: calorimetric tests, X-ray diffraction, scanning electron microscopy, mercury intrusion porosimetry, and compressive strength tests. To avoid the potential difficulties related with the increase in the solution volume, an alternative mix design is proposed, which can produce concrete with mechanical properties—including the compressive strength—that are equal to or better than the traditional control mix, while simultaneously preserving the outstanding temperature reduction capabilities of urea.

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1. Introduction

Urea ($\text{CO}(\text{NH}_2)_2$) is an organic compound widely used in physiological processes and in nitrogen fertilizer related research. Lewis and Burrows [1] studied the equilibrium between urea–water and carbon dioxide–ammonia, and proved that free energy was transferred through the reaction.



The forward reaction in (1) (urea reacts with water to produce a solution) is endothermic and reduces the ambient temperature, while the reverse reaction (ammonia and carbon dioxide react to produce urea) is exothermic and releases energy [2,3]. The reaction equation (1) expresses only thermodynamic transfer; therefore, the actual reaction results may differ from it. This heat transfer of urea is identical to that of the phase change materials (PCMs) that are typically used to reduce the hydration heat of cement in mass concrete. Phase change materials absorb or release heat through phase changes; as latent heat storage systems, they present different latent heat and transition temperatures [4]. Many

studies were performed on the use of the latent heat of PCMs to improve the thermal properties of mass concrete, or to reduce the temperature of concrete in regions with high temperatures, such as in tropical environments [5–7].

The set of PCMs generally used with concrete can be divided into three categories: organic, inorganic, and eutectics [8,9]. Aside from eutectics—for whose thermo-physical properties there is no adequate test method—organic PCMs are known to be preferable to inorganic PCMs [10], because they exhibit useful advantages and their disadvantages are manageable.

The urea used in this study—whose adequacy as an additive to concrete is to be investigated—is an organic and eco-friendly material. Urea is mainly comprised of carbon dioxide and ammonia, both of which have been categorized as air pollutants. Therefore, adopting urea in industries with high usage and demand such as the concrete industry will increase the consumption rate of carbon dioxide in the atmosphere. However, urea is still expensive in some countries—including South Korea—where CO_2 capture technology and storage facilities have not yet been established; in such cases, using urea as a concrete additive can increase the price of concrete. For this reason, the immediate application of urea is difficult in practice. Nevertheless, the use of urea in the concrete industry in

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South Korea (which has an annual demand of approximately 150 million m³) is expected to bring significant ecological benefits.

Aside from the environmental advantages, another advantage of mixing urea in concrete is the effective reduction in the hydration heat of cement. When it transitions from the solid phase to the liquid phase, urea can absorb more heat than other PCMs [2,11]. The key point is that the melting point of urea is within the ambient temperature range. Consequently, when urea is mixed with fresh concrete, its instant dissolution in the mixing water will significantly decrease the temperature of concrete. The resulting effect is similar to that of the conventional method of cooling each individual material before mixing, in order to reduce the temperature of concrete in hot weather conditions [12].

Although much research has been conducted on organic PCMs [5,7], not many studies have addressed the use of urea as an additive to concrete. Applying urea as an additive to concrete was expensive, because it was already being used in the agricultural industry as a promoter of plant growth or as a crop fertilizer. In addition, the full appreciation (by the concrete industry) of the advantages of PCMs for stabilizing the thermal properties of mass concrete is quite recent.

The purpose of this study is to investigate various properties of urea, including heat absorption in cement composites. A typical cement composite was fabricated with no urea content (0%), to be used as a control mix. Several cement composites were then prepared with step increases in the urea mix ratio. Four different urea mix ratios were thus obtained as binder (0, 5, 10, and 15% by mass), in order to study the solution temperature—that is, the cement composite temperature—decrease with the increase in the concentration of urea dissolved in water. It was anticipated that an increase in the urea concentration would also lead to an increased reduction of the hydration heat of cement. In addition, high volumes of other supplementary cementitious materials (SCMs) were used, in order to compare their hydration heat reducing capacities. The SCM method decreases the amount of ordinary Portland cement (OPC) and replaces it with by-products used as binders, such as fly ash (FA) and ground granulated blast-furnace slag (GGBS). This method has been used in many studies in order to reduce the hydration heat [13,14]. This study aims to perform a comprehensive evaluation of the effects of the use of urea as an alternative for decreasing the hydration heat, in what concerns its impact on the thermal, mechanical, and pore structure properties of the resulting cement composites.

2. Experimental approach

2.1. Experimental setup

The anticipated reaction from using urea as an additive was the following. First, the urea would dissolve in water immediately after being added, as a result of forced mixing. The resulting endothermic reaction would drastically reduce the temperature of the cement composite and, as a result, the lowered temperature would reduce the peak of hydration heat from the cement.

As a first step, this study evaluated the heat absorption properties of urea, according to the different urea solution concentrations; the capability of urea to mitigate the heat generated during the exothermic cement hydration was also examined. The experiment therefore began by simply dissolving urea in water. A paste test was then conducted by adding cement to the solution, which was followed by the addition of sand, to conduct a mortar test.

The increase in strength of low-heat concrete—which is mainly used in mass concrete or in hot weather conditions—occurs very slowly in typical temperature environments, because of its low

hydration heat. Furthermore, when urea is mixed into the concrete, not only is the temperature decreased, but the volume of solution is also increased (by the volume of urea dissolved in water). Once hydration is complete, the urea remaining in the paste may increase the porosity of cement hydrates. The joint effect of the low hydration heat and the high porosity will hinder the cohesiveness of cement and may decrease the strength of the resulting concrete.

Table 1 shows the mix ratios of the cement composites used in the experiment. The urea mix ratios for the binder mixes were each set to 5, 10, and 15% by mass. The water to binder ratio (W/B) of the cement composites was fixed at 0.45 by mass for all the mixes.

Table 1 shows a noteworthy result: the increase in urea ratio in the mixes was accompanied by an increase in the urea solution (urea and water) to binder volume ratio, even though the water to binder ratio was maintained fixed at 0.45. This resulted from the fact that the volume of urea solution was increased by the volume of urea that dissolved in water from the forceful mixing of the cement composite. However, the urea dissolved in water should not affect the hydration of cement; therefore, in theory, the W/B ratio required for binder hydration should not increase.

Typically, when the water volume increases for a fixed quantity of binder (for example, when the W/B increases), the increase is felt only on the volume of residual water—the water that did not participate in the hydration of cement. This decreases the compressive and flexural strength of the cement composite. The observed increase in the urea solution to binder volume ratio with the increasing urea ratio is a similar event. Consequently, the impact of the urea mix ratio on the resulting strength and porosity characteristics of the cement composite needed to be verified. To verify the dependence of the mechanical properties of the cement composite on the solution volumes, the test series designed for mortar (Series II) used a mix in which the water volume was reduced by the volume of urea dissolved in water (Ur4). The other mixes replaced the volumes of binders, water, and sand by the same ratio of mixed urea, whereas Ur4 only replaced water; the urea solution to binder volume ratio in this sample was therefore equal to that of the control mortar (Co). For the mortar tests (Series II), the volume of mixed urea was 63 L/m³ for both Ur3 and Ur4, but the solution volumes showed differences of 46 L/m³; this is because in Ur4 the water volume was reduced by the full amount of added urea. The amount of heat absorbed by urea was therefore expected to be similar in both cases, but differences in strength and pore size distribution were expected. These differences should allow a comparison of the mechanical properties of the resulting cement composites.

2.2. Methods

The hydration heat of the urea-blended pastes was measured. The heat flow was monitored during a 72 h isothermal calorimetric test, and the resulting data were integrated (to obtain the cumulative heat) and analyzed (to determine the effect of urea on the early hydration). A twin-conduction type calorimeter manufactured by Tokyo Riko was used. The samples were prepared by placing the binder and urea in separate sample cups in the calorimeter, and equilibrating them to 23 °C. The water used for cement hydration and urea dissolution was also equilibrated to 23 °C in the calorimeter before being injected into the prepared urea and cement powder. Finally, the acquisition system was started, to record the heat evolution rate during the first 72 h of hydration.

X-ray diffraction (XRD) analysis was also performed at the age of 3 d. An X-ray diffractometer (D/MAX-2500) manufactured by Rigaku was used for analyzing the chemical variations. The maximum power of the X-ray generator was 60 kV; data was collected between 5 and 70° (2θ range) with a scan speed of 14 s per step.

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