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Numerical Model for Quantifying Degree of Hydration in Concrete Mixes with Reduced CO2 Footprint

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Abstract

The widespread application of innovative cementitious combinations in concrete raises the need for more comprehensive investigation of the resulting concrete properties. Early age behaviour is a major factor to be addressed, and tools are required for quantifying the hydration state of concrete members, particularly at early-ages. Numerical models can potentially be used in mass concrete construction to predict and prevent possible thermal crack formation. They also provide an indirect means for characterizing development of the hydration reaction in concrete. The latter can then be utilised in modelling and predicting secondary concrete properties, such as diffusion coefficient. This is gaining increasing importance as we harness the ability to develop innovative combinations.

The cement industry is estimated to be responsible for about 7% of the carbon dioxide generated globally. As such, reducing the amount of CO_2 emitted during cement production is a key issue if the construction industry is to fully participate in sustainable development. Under the terms of the Kyoto Protocol Emissions Trading Scheme it is also potentially profitable for cement companies to reduce their CO_2 emissions. By using blended cement instead of ordinary Portland cement, it is possible to lower the share of clinker in cement, resulting in reduced CO_2 and energy emissions. In Ireland, CEM II now accounts for over 80% of the Irish cement production portfolio.

GGBS is a by-product of steel industry and a common replacement for cement. When compared to Portland cement it has a reduced CO_2 footprint and concretes containing GGBS are less prone to deterioration due to aggressive chemical attacks. Its use has the potential to produce more durable concrete with increased service life, lower maintenance costs and a lower carbon footprint, increasing the sustainability of concrete construction.

The aim of the current study is to use numerical models to quantify the development of heat of hydration when mixtures of CEM II and GGBS are utilised. Experiments were conducted where the temperature profiles in 4 different mixes of concrete (CEM II with 0%, 30%, 50% and 70% GGBS) are recorded. This was achieved by casting 6 identical concrete samples from each mix, with thermocouples embedded to record the internal temperature of the mix at regular time steps. Temperature changes of the mix are then used to quantify the heat evolved, based on the principles of heat transfer. To account for the combined effect of time and temperature on hydration development, activation energy of the mix is used, along with the equivalent age maturity method. Total heat of hydration is determined based on the composition and amount of cementitious materials.

It has long been accepted that the liberated heat of hydration, divided by the total available heat of hydration is a good measure of the degree of hydration. The experimental data describing hydration development with equivalent age are then used to calibrate the exponential formulation presenting the S-shaped hydration curve. Values of β, τ, and α_u (the hydration parameters) are obtained for each mix, from the results of multivariate non-linear regression analysis. Comments on the use of this method in quantifying concrete hydration are then made.

1. Introduction

Concrete, with estimations of more than 10 billion tons production worldwide, has an enormous impact on the environment, whether through the natural resources it consumes as the raw material, or through the energy consumption and the huge amount of CO_2 released as a result of cement production [1]. It has been estimated that the cement industry is responsible for about 7% the total CO_2 generated worldwide [2]. Reducing this figure is of great importance for the cement industry to contribute significantly in the construction sector's contribution to global sustainable development.

The key ingredient in production of Portland cement is cement clinker. It is an intermediate product, resulting from calcination of limestone and subsequent fusion with clay minerals. The clinker is then removed from the kiln to cool, ground to a fine powder, and mixed with a small fraction (about five percent) of gypsum to create the most common form of cement known as Portland cement [3]. The reaction of converting limestone to lime, involves the release of large quantities of CO_2:

$$CaCO_3 + \text{Heat} \rightarrow CaO + CO_2$$

Moreover, the cement manufacturing process requires that materials be heated to temperatures in excess of 1400^oC to achieve full fusion between the lime and clay minerals. If the carbon footprint resulting from this heating is also taken into account, the production of one tonne of Portland cement leads to the release of approximately 0.95 tonnes of CO_2 [4].

Since 1995, the cement industry has committed itself to decreasing their CO_2 emissions, in order to contribute to sustainability and global warming prevention [5]. However, the limited ability to reduce CO_2 emissions in manufacturing ordinary Portland cement necessitates the development of alternative cement binders. One approach to do this is to replace OPC with blended cements in order to lower the share of clinker in the final product [6]. Additions such as ground granulated blastfurnace slag (GGBS) -which is a by-product of steel industry- or fly ash, can be mixed with clinker to produce blended cements. Since these additions reduce the overall demand for clinker, they will result in more environmental friendly cements. They have already been in use across the Europe for several years, though their application depends on the local availability of the clinker substitutes. The percentage replacement of clinker can vary from 5% up to typically 70% (although higher replacement values are technically possible). In effect, the lower the share of clinker, the lower the CO_2 emission associated with the cement [7].

Blastfurnace slag (BS), a by product of the steel industry if quenched by water forms a glassy material known as granulated blastfurnace slag (GBS), which can show hydraulic properties when in contact with water. Grinding this material to a fine powder (GGBS) enhances it properties as a cement replacement material. The rate of its reaction with water is slow; however, when mixed with Portland cement, the alkalis and sulphates released during cement hydration can act as activators to raise the hydration rate of GGBS [8]. Concrete containing GGBS has a higher proportion of calcium silicate hydrates (CSH), the ingredient that contributes to the concrete strength. This leads to production of concrete with a higher ultimate strength compared to the concrete made with OPC. Moreover, this type of concrete continues to gain strength over time, and has been shown to double its 28 day strength over periods of 10 to 12 years [9]. GGBS also enhances the durability and resistance of concrete against aggressive environments. It can be used in large volume concrete pours to limit excessive temperature rises. It causes the heat to be generated more gradually, with lower peaks and less total heat of hydration, limiting thermal gradients in concrete. Excessive thermal gradients can lead to formation of micro-cracks and consequent reduction in durability.

Constituents such as GGBS -also known as Secondary Cementitious Materials- can either be used in combination with other cements in the concrete at time of mixing, or can be used as a partial replacement for clinker during the cement production process to produce a single-powder 'blended cement'. Blended cements involving high levels of GGBS replacement can lead to long-term performance enhancement in aggressive environments, compared to Portland cement alone [8]. They are suitable in most applications; however, consideration must be given to the possibility that other characteristics of the concrete might be affected as a result, e.g. initial strength, drying time, resistance to seawater attack [7]. Widespread application of innovative cementitious combinations underscores the need for more close investigations of properties of the resulting concrete. One of the important fields to be addressed is the early-age behaviour of concrete. To be able to develop a model that predicts the in-place early-age and even long-term performance of concrete, an accurate estimate of the hydration development of the mix should be made [10]. This development can be characterised by the 'degree of hydration' and by hydration curves based on three key parameters.

The degree of hydration (α) is a measure that quantifies how far the reactions between water and cementitious materials have progressed. It is defined as the ratio between the quantity of hydrated cement grains and the original quantity of cement grains available in the mix. Hydration degree increases with age and concrete maturity. The curing temperature of the mix is also another influential parameter. Different approaches have been proposed to quantify the degree of hydration of concrete mixes. Some are direct approaches, aimed at measuring the quantity of cement gel formed in the mix, while others are indirect methods, estimating the degree of hydration based on the amount of chemically-bound water or the amount of heat released during hydration [11]. The indirect method based on the released heat of hydration has been adopted in this study to develop the hydration curves. The hydration curve can then be used in characterization of the hydration behaviour of concrete mixes at a specific curing temperature, known as the reference temperature (T_r). Temperature sensitivity of the mix and the equivalent age maturity method can then be employed to predict the behaviour under various curing temperatures encountered in practice.

Therefore, numerical models are sought to characterize the hydration development of concrete specimens in their early-age. These models can then be implemented in predicting related concrete properties, such as permeability and diffusivity. These properties are influenced by pore size distribution and the structural formation of crystals within the paste, which in turn, are affected by hydration development and the rate of heat generation. Numerical models can also be used in predicting early-age strength gain of concrete and estimating the amount of heat generated during hydration. Knowledge of the heat release patterns is essential in large concrete pours, to avoid high temperatures and excessive thermal gradients, which can lead to durability problems in the resulting concrete.

Currently, limited guidance is available for characterizing the hydration behaviour of mixes, especially when Secondary Cementitious Materials are used. The purpose of this study is to contribute to addressing this gap in the guidance available. This is achieved by determining hydration curve parameters through an experimental study of concrete mixes made using CEM II binders with varying replacement levels of GGBS.

2. Experimental programme: materials and methods

2. 1. Materials

A laboratory program was designed to monitor the heat generated during hydration of four different concrete mixes, comprising limestone aggregates, CEM II A-LL (Irish source) and various replacement levels of GGBS, ranging from 0 to 70%. Table 1 provides a summary of the concrete mixtures tested. Hydration characterization curves were derived from measured data and use of published methodologies for determining the degree of hydration and the 'equivalent age' concept.

From each concrete mix, six cubic specimens, 30*30*15 cm each, were cast. Embedded thermocouples were used to record the internal temperature of specimens. Heat of hydration was then calculated for each mix, based on the average values recorded in these samples. Temperature monitoring was carried out for several days after batching, until the temperature stabilised. To prevent excessive heat loss to the environment, insulation boards, 2 cm thick were used to seal off the concrete specimens while they are being cured.

Once the degree of hydration development and equivalent age of specimens were determined using the experimental data, multivariate non-linear regression analyses were performed, to obtain the corresponding values of hydration curve parameters for each mix

Table 1: Material proportions for the four different mixes used in the experiment.

Mix	GGBS level (%)	Cement Content (kg/m^3)	GGBS Content (kg/m^3)	Fine aggregate (kg/m^3)	C10 aggregate (kg/m^3)	C20 Aggregate (kg/m3)	Water Content (kg/m^3)
1	0	320	0	970	325	945	185
2	30	225	95	970	325	945	205
3	50	160	160	970	325	945	195
4	70	95	225	970	325	945	195

2. 2. Methodology for quantifying the degree of hydration based on the released heat

It has been widely accepted that the cumulative amount of heat evolved at time t of the hydration process, divided by the total amount of heat available at 100% hydration, can be used as the measure of degree of hydration [11, 12]:

$$\alpha(t) = \frac{H(t)}{H_T} \quad (1)$$

where

$\alpha(t)$: degree of hydration at time t

$H(t)$: cumulative heat generated from time 0 to time t (J/g)

H_T: Total heat generated assuming complete hydration (J/g)

Cements have various chemical compositions, and each of their constituents has been found to have a unique heat of hydration [10]. Once the percentage of each phase is known in the cement paste (e.g. using Bogue formulations) the total heat of hydration of cement at full hydration (H_{cem}) can be quantified using equation 2.

$$H_{cem} = 500p_{C3S} + 260p_{C2S} + 866p_{C3A} + 420p_{C4AF} + 624p_{SO3} + 1186p_{FreeCaO} + 850p_{MgO} \quad (2)$$

where

H_{cem}: total heat of hydration of cement (J/g)

p_i: weight ratio of the i-th compound in terms of total cement content

In the current study, the concrete mix is a blend of cement and GGBS. To quantify the total heat of hydration of the cementitious system, the heat of hydration of GGBS is also required, and based on previous research it has been considered to be 461 J/g [13]. Equation 3 can then be used to calculate the total heat of hydration of cementitious system, when a mixture of cement and GGBS is used:

$$H_u = H_{cem} p_{cem} + 461 p_{slag} \quad (3)$$

where

H_u: total heat of hydration of cementitious material (J/g)

p_i: weight ratio of the i-th component in terms of total cement cementitious materials

With knowledge of the total cementitious materials content per unit volume of concrete (c_c), the ultimate heat of hydration at 100% hydration can be calculated using Equation 4:

$$H_T = H_u c_c \quad (4)$$

where

H_T: total ultimate heat of hydration of cement (J/m^3)

c_c: cementitious materials content in unit volume of concrete (g/m^3)

2. 3. Methodology for Characterizing the Hydration Behaviour Curves

Once the evolved heat of hydration of the concrete mixes has been experimentally determined, and the degree of hydration development estimated, a best-fit mathematical model can be proposed to represent the data [11]. In this current study, the exponential formulation presented in equation 5 has been used. It is shown to accurately represent the s-shape of the hydration development curve [14, 15]:

$$\alpha(t_e) = \alpha_u . \exp(-[\frac{\tau}{t_e}]^{\beta}) \quad (5)$$

where

t_e: equivalent age at the reference temperature (T_r)

α_u: ultimate degree of hydration

β: hydration shape parameter

τ: hydration time parameter (hours)

The hydration curve parameters (α_u, β, τ) represent the amount of acceleration, retardation, rate and ultimate degree of hydration in different mixes, with τ being relevant to the timing of the accelerating part, and β indicating the rate of hydration [12]. In practice, the hydration process almost always stops before the cement is totally consumed, and a degree of hydration of 100% may never be reached [17]. α_u has been introduced to the equation to allow for this effect to be considered in the hydration curve mathematical model. This variable is strongly affected by the water-cement ratio of the mix, though it remains unaffected by the curing temperature [16 - 18].

The other parameter in equation 5 (t_e), represents the equivalent age of a specimen cured at the reference temperature (T_r). Curing temperature has the most significant effect on the rate of hydration [19, 20]. Therefore, to determine maturity of a concrete specimen, curing temperature should also be taken into account, along with the chronologic curing age. To be able to account for the combined effect of time and temperature, the maturity method and equivalent age function developed by Freiesleben, Hansen and Pedersen [19] is used. This non-linear function (equation 6) converts the actual age (t) of a concrete specimen cured at any temperature (T_c) to an equivalent curing age (t_e), assuming the sample has been cured at the reference temperature (T_r) [20]:

$$t_e(T_r) = \sum_0^t \exp(\frac{E}{R}(\frac{1}{273+T_r} - \frac{1}{273+T_c})).\Delta t \quad (6)$$

where

$t_e(T_r)$: equivalent age at the reference curing temperature (h)

Δt: chronological time interval (h)

T_c: average concrete temperature during the time interval Δt (˚C)

T_r: reference temperature (˚C)

E: activation energy (J/mol)

R: universal gas constant, 8.3144 J/mol/K

In equation 6, 'activation energy' (E) indicates the temperature sensitivity of a concrete mixture [10]. Several formulations have been proposed to calculate this parameter for a mix. The model developed by Schindler [11] is employed here:

$$E = 22{,}100 f_E \, (p_{C3A})^{0.3} \, (p_{C4AF})^{0.25} \, \text{Blaine}^{0.35} \quad (7)$$

$$f_E = 1 + 0.4 \, p_{slag}$$

where

p_i: weight ratio of the i-th compound in terms of total cement content

Blaine: specific surface area of cement (m^2/kg)

f_E: activation energy modification factor when GGBS is added to the mix

2. 4. Methodology for quantifying the heat generation of the mix

By monitoring the temperature development of a concrete mixture during hydration, the amount of heat generated can be quantified. Unless the concrete specimens are cured in a fully-adiabatic condition, there is always an amount of heat-loss in the system, and temperature development is not as high as the adiabatic temperature rise. This amount of heat-loss should also be considered, when calculating the total heat evolved in the system, up to the time t, to give us the value of H(t) to be used in equation 1.

The heat generated due to cement hydration raises the internal temperature of concrete specimens. During the first days after casting, the rate of hydration is higher and results in a considerable temperature build-up in the specimens. The difference between concrete and ambient temperature will result in a heat flow to the environment, which can be quantified based on the laws of heat transfer. Here, the one-dimensional heat diffusion equation (equation 8) has been used [21]:

$$-Q = \rho c A \frac{d(T_c - T_{amb})}{dt} \delta x \quad (8)$$

where

Q: the rate of heat loss (J/h)

ρ: density of the insulation slab (kg/m^3)

c: specific heat capacity of the slab (J/kg/K)

A: surface area of the insulation slab (m^2)

δx: thickness of the insulation board (m)

T_c: internal temperature of the concrete samples (˚C)

T_{amb}: ambient temperature (˚C)

t: time (h)

The net amount of heat captured inside the specimens, responsible for raising the internal temperature of concrete can also be quantified using equation 9 [11]:

$$\frac{dH}{dt} = \frac{dT}{dt} \rho c_p \quad (9)$$

Specific heat of concrete is one of the parameters that is used in quantifying the amount of heat generated in the hydration process. This parameter does not remain constant during the early-age of concrete, since it is highly influenced by the amount of unbound water content in the mix, which decreases over time, as the hydration degree progresses [22]. The following equation has been employed here to account for the changes in specific heat capacity of concrete in early-age [23]:

$$c_p = \frac{1}{\rho}(W_c.\alpha.c_{cef} + W_c.(1-\alpha).c_c + W_a.c_a + W_w.c_w) \quad (10)$$

$$c_{cef} = 8.4T_C + 339$$

where

c_p: specific heat capacity of concrete (J/kg/K)

ρ: density of the insulation slab (kg/m^3)

$W_{c,a,w}$: weight ratio of cement, aggregate and water in concrete mix (kg/m^3)

$c_{c,a,w}$: specific heat of cement, aggregate and water (J/kg/K)

c_{cef}: specific heat of hydrated cement (J/kg/K)

α: degree of hydration

T_c: concrete temperature (˚C)

Using this approach, the amount of heat loss, and the net heat responsible for temperature rise of concrete can both be quantified at discrete times after batching. The sum of these two values is the total heat generated in the hydration process, H(t), to be used in equation 1.

3. Results

Figure 1 summarizes the results obtained during temperature monitoring of the mixes (measured concrete temperatures), along with the adiabatic temperature development, calculated based on the laws of heat transfer. It may be seen that, as expected, the peak temperature drops with increasing levels of GGBS but the profile of the cumulative heat generated over time increases with increasing levels of GGBS. The results show that the maximum temperatures reached during hydration decrease with increased.

Figure 2 represents the heat evolution trends noticed in the mixes. These are the diagrams of the total heat released by each mix, during their hydration process, calculated based on the laws of heat transfer, and using the temperatures recorded over the first week of monitoring the specimens. The parameters which determined the s-shape hydration curves, based on the results of regression analyses, are shown in Table 2.

Table 2: Hydration parameters obtained from regression analysis of experimental data (T_r = 21.1°C)

No	Mix Description	E (J/mol)	H_u (J/g)	α_u	τ	β
1	CEM II	45105	474.5	0.417	4.266	1.503
2	CEM II + 30% GGBS	50518	470.5	0.379	5.101	1.170
3	CEM II + 50% GGBS	54127	467.8	0.369	6.906	0.575
4	CEM II + 70% GGBS	57735	465.1	0.312	8.714	0.606

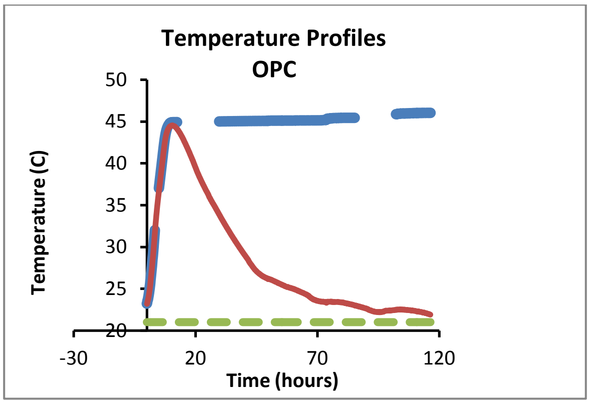

Figure 1- a

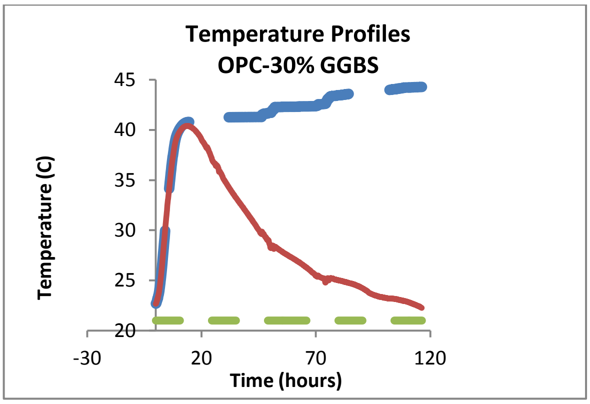

Figure 1- b

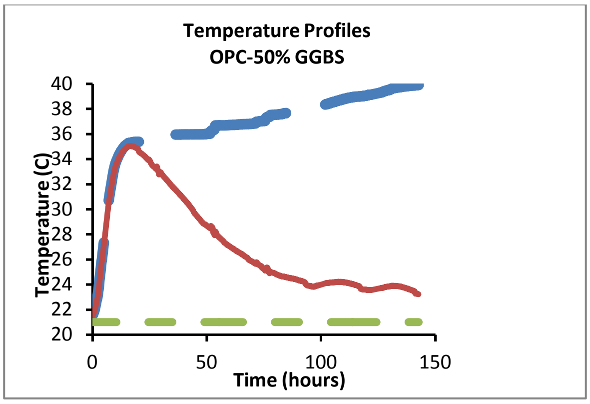

Figure 1- c

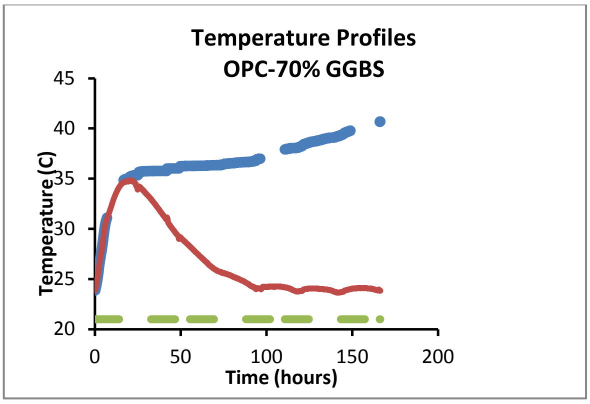

Figure 1- d

Figure 1: Measured semi-adiabatic vs. ideal temperature rise (assuming no heat loss)

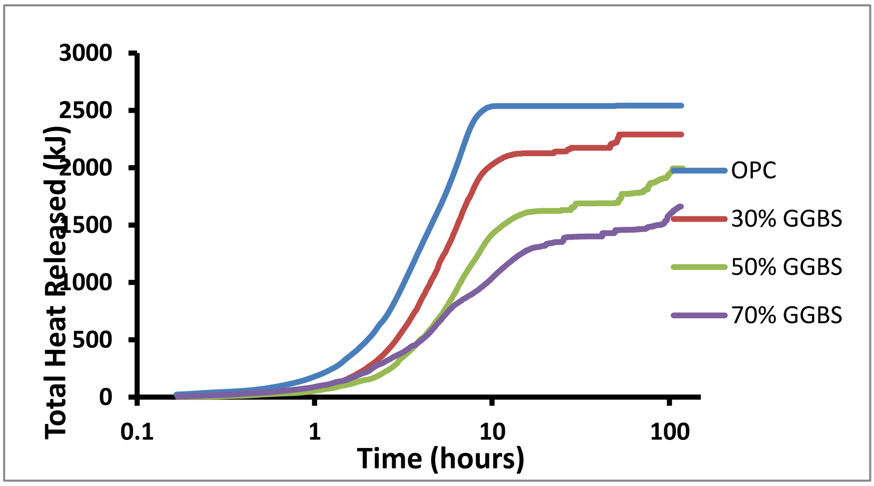

Figure 2: Profile of the total heat released during the hydration process of the 4 mixes

The hydration characterization curves, produced by substituting the values of hydration curve parameters for each mix (given in table 2) in equation 5 are shown in Figure 3, and can be utilised in predicting the degree of hydration development of the mixes.

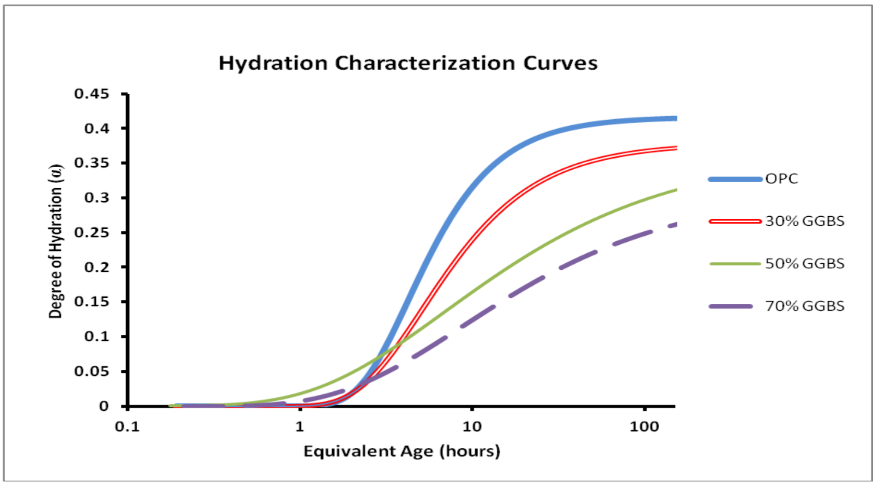

Figure 3: S-shaped hydration characterization curves

4. Discussion

As it can be seen in the results, increasing the amount of GGBS delays the start of the acceleration stage. This is reflected in the increasing hydration time parameters (τ), which double in value as the cement replacement level goes from 0% to 70%. The rate of hydration development, (the slope of hydration curve during the acceleration stage) slows down as the percentage level of GGBS increases in the mix. As a result, the values obtained for the hydration rate parameter (β in table 2) reduce by a factor of 2 as the cement replacement level goes from 0% to 70%. Equally, a trend is observed whereby the total heat released by the end of temperature monitoring is decreasing, which results in the values calculated for α_u to decrease from 0.4167 to 0.3122.

While considering the equations proposed in the literature for estimating the values of this parameter, increasing the amount of GGBS in the mix, should in fact increase the ultimate degree of hydration of the resulting concrete [10]. This can be attributed to the fact that the rate of heat generation slows down considerably in mixes with higher levels of GGBS (50% and more). This might imply that the duration of temperature monitoring considered in this study (1 week) has not been long enough for the hydration process in a GGBS containing mixture to reach degrees of hydration close to their actual α_u. This hypothesis can be underscored by the fact that by the end of the first week after batching, the recorded internal temperatures of concrete had not stabilized with the ambient temperature. Also, in Figure 3, comparison of the hydration characterization curves obtained for different mixes shows that in the case of the mix with 70% GGBS, the s-shape hydration curve has not been completely formed by the end of the 1st week. Considering all of these, one may suggest that in order to investigate the hydration of mixtures containing high levels of GGBS, the temperature monitoring period should be extended beyond 1 week.

In order to address this, and to be able to obtain more precise estimates of α_u, another solution would be to adopt the more direct approach for determining the degree of hydration, rather than using the heat of hydration development. Another study is currently being carried out to investigate this. In the new study the degree of hydration will be evaluated based on analysis of back-scattered electron microscope images taken from concrete specimens at different ages.

5. Conclusions

Comparison of the values obtained for hydration curve parameters (Table 2) with those reported in the literature for mixtures based on CEM I, shows a considerable difference, which reflects the significantly different hydration behaviour of cement/addition combinations.

This emphasises the need for further validation studies of the application of numerical models based on the parameters which determine hydration curves.

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