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ORIGINAL PAPER

ANALYSIS OF THE HYDRATION PROCESS OF CEM II/B-M COMPOSITE CEMENT

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Abstract. *This paper presents the hydration kinetics of a CEM II/B-M type Portland composite cement by DTA/TGA analyses and mathematical and software modeling prediction. The reason for this choice is generated by its use as the main binder in the concrete matrix used for radioactive waste conditioning processes and the lack of public information related to its hydration process. The comparison between the experimental and modeling results indicated a good agreement and correlation of the results, reinforcing the idea that modeling can be a powerful tool that can be used in understanding this complex multicomponent-multiphase system of hydrated cement and can facilitate the assessment of the geochemical evolution of cement-based materials under different environmental conditions.*

Keywords: *cement hydration; software modelling; DTA/TGA analyses; Portlandite; chemically bound water*

1. INTRODUCTION

Materials based on Portland cement are widely used due to the properties they possess and the low production costs. According to European Standard EN 197-1 [1] there are 27 standard cements classified into five main types, from CEM I to CEM V. For this study it was chose CEM II type, subtype B-M (S-LL). The reason for this choice is generated by its use as the main binder in the concrete matrix used for radioactive waste conditioning processes.

In the context of their use in special fields such as nuclear applications, these materials must be given special importance from the point of view of their stability and durability for a long period (ten thousand to hundred thousand years). Understanding cement-based materials begins with understanding the first step that leads to their formation, namely the hydration process.

Hydration of cement represents its reaction at contact with water, after which hydration products such as CSH (Calcium Silicate Hydrate), CH (Portlandite), $C_6A\bar{S}_3H_{32}$ (Ettringite), $C_4A\bar{S}H_{12}$ (Monosulfate) and others are formed. Hydration is a very complex process that involves a lot of chemical reactions directly influenced by the temperature and the energy of the system.

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During the last few years, many papers [2-8] reported models and methods which applied to cementitious systems estimate the degree of hydration and phase assemblage. Most of them are based on experimental approaches such as X-ray diffraction (XRD), thermogravimetric analysis (TGA) and scanning electron microscopy- backscattered electrons (SEM-BSE) and/or software modeling using geochemical codes such as GEMS developed by PSI (Paul Scherrer Institute) [9] and PREEQCH developed by USGS (U.S. Geological Survey) [10].

It has been found only a small number of publications [11, 12] that reported studies related to the hydration of CEM II/B-M type Portland composite cement and its field of use. With this in mind, both work approaches (experimental determination and software modeling) were taken into account in order to build a valid kinetic hydration model.

A full understanding of the hydration process is crucial for future assessment of cement-based materials' performance, because its main properties such as workability, setting behaviour, strength development, but also durability, are influenced to this process [13].

2. MATERIALS AND METHODS

2.1. MATERIALS

The cement CEM II/B-M (S-LL) type used in this study is a commercial Portland composite which according to EN 197-1 [1] has in its general composition 65-79% clinker and 21-35% additional materials (in this case slag and limestone).

The oxide proportions and normative phase composition are shown in Table 1. The oxide proportion was taken from the technical data sheet of the product. The normative phase was obtained using modified Bogue's Equations of Stutzman et al. [14] for the determination of the oxide component and modified Taylor's Models of Shim et al. [15] for the determination of the sulfate component. Also, Loss on ignition (LOI) and CO_2 was determined experimentally by thermal analysis.

Table 1.Oxide proportions and normative phase composition of the cement

Chemical composition	[%]	Normative phase composition	[%]
CaO	59.7	Alite (C_3S)	22.085
SiO_2	21.4	Belite (C_2S)	39.843
Al_2O_3	5.7	Aluminate (C_3A)	8.625
Fe_2O_3	3.4	Ferrite (C_4AF)	9.538
CaO (free)	0.5	CaO (free)	0.461
MgO	1.41	$CaCO_3$	13.838
K_2O	1.1	$CaSO_4$	4.308
Na_2O	0.2	K_2SO_4	1.14
CO_2	6.2	Na_2SO_4	0.09
SO_3	3.4	K_2O	0.25
(LOI)	7.1	Na_2O	0.07
Blaine surface area [m^2/kg]	432	MgO	1.3
Total Additive	27	SO_3	0.09

2.2. METHODS AND EXPERIMENTAL SETUP

2.2.1. Determination of hydration degree by thermal analysis

Thermal analysis is an experimental method that can be used to estimate the degree of hydration of a cement paste by coupling two techniques: differential thermal analysis (DTA) and thermogravimetric analysis (TGA). DTA locates the ranges corresponding to thermal decompositions of different phases in the paste by comparison with a reference material, while TGA simultaneously measures the weight loss due to the decompositions as a function of temperature and/or time [7, 16].

The DTA/TGA analyses were conducted on hardened cement pastes prepared with a 1/2 water to cement ratio (w/b) at room temperature (22°C±2). After mixing the cement paste was cast in sealed tubes until the age of testing to avoid water evaporation and air carbonation. No method of stopping hydration was used, thus preventing carbonation [5]. The cement pastes were analysed by heating them from room temperature to 1000°C in a nitrogen atmosphere immediately upon reaching the testing age.

There are several methods to estimate the degree of cement hydration using DTA/TGA diagrams. These methods are based either on the quantification of Portlandite (CH) or chemically bound water (W_b).

To determine the total amount of Portlandite and chemically bound water generated during hydration Chu et al. [5] propose to use the following simplified equations:

$$C_3S + 5.3H -> C_{1.7}SH_4 + 1.3CH \tag{1}$$

$$C_2S + 4.3H -> C_{1.7}SH_4 + 0.3CH \tag{2}$$

$$C\bar{S} + 2H -> C\bar{S}H_2 \tag{3}$$

$$C_2A + 6H -> C_3AH_6 \tag{4}$$

$$C_3A + 3C\bar{S}H_2 + 26H -> C_6A\bar{S}H_{32} \tag{5}$$

$$C_3AF + 3C\bar{S}H_2 + 30H -> C_6A\bar{S}H_{32} + CH + FH_3 \tag{6}$$

$$2C_3A + C_6A\bar{S}H_{32} + 4H -> 3C_4A\bar{S}H_{12} \tag{7}$$

$$2C_3AF + C_6A\bar{S}H_{32} + 12H -> 3C_4A\bar{S}H_{12} + CH + FH_3 \tag{8}$$

In the study there were used three methods for estimation of the hydration degree of cement using DTA/TGA diagrames. Two of them are based on Portlandite quantification (Method 1 and Method 3) and the third one (Method 2) is based on chemically bound water quantification.

Method 1

The first method used for estimating the degree of hydration is based on CH quantification by using Equation (9) proposed by Duc Chinh Chu *at al* [5].

$$\alpha(\mathrm{t}) = \frac{m_{CH}}{m_{C}\mathrm{r}_0} * 100 \tag{9}$$

where:

α(t)- degree of hydration of the cement at time t;

$m_{Ca(OH)_2}$- amount of CH calculated using the Equation (10);
m_C- mass of the initial cement calculated using the Equation (11);
Γ_0- amount of CH produced upon complete hydration of the cement.

$$m_{CH} = \frac{\Delta m(420°\mathrm{C} - 540°\mathrm{C}) * M_{CH}}{M_{H_2O}} \tag{10}$$

where:

$\Delta m(420°\mathrm{C} - 540°\mathrm{C})$- the mass loss of the samples between 420°C and 540°C;
M_{CH}- molar mass of CH;
M_{H_2O}- molar mass of water.

$$m_c = \frac{m_{sample}}{\left(1 + \frac{w}{c}\right)(1 + LOI)} \tag{11}$$

where:

m_{sample}- mass of the samples;
w/c- the water to cement ratio of the paste;
LOI- loss of cement on ignition.

Method 2

The second experimental method of estimating the degree of hydration is to quantify the chemically bound water and use it in the following equation [9, 12]:

$$\alpha(\mathrm{t}) = \frac{W_b(t)}{W_b(\infty)} * 100 \tag{12}$$

where:

$\alpha(\mathrm{t})$- degree of hydration of the sample at time t;
$W_b(t)$- amount of bound water for a sample at time *t* which can be determined experimentally by Equation (13);
$W_b(\infty)$- amount of water bound for a completely hydrated.

$$W_b(t) = \frac{\Delta m(105°C - 1000°C)(t)}{m_{sample}1000°C(t)} * 100 \tag{13}$$

Method 3

The third method uses Equation (14) to estimate the degree of hydration and takes into consideration quantifications of portlandite both at a certain point of hydration and at the end of hydration. Is a simpler form of Equation (9) and was adapted from Wang et al. (2020) [18] and Chu et al. (2021) [5].

$$\alpha(\mathrm{t}) = \frac{m_{CH}}{m_{CH_{(\infty)}}} * 100 \tag{14}$$

where:

$\alpha(\mathrm{t})$- degree of hydration of the sample at time t;
m_{CH}- amount of $Ca(OH)_2$ calculated using the Equation (10);
$m_{CH_{(\infty)}}$- the $Ca(OH)_2$ content in a fully hydrated cement paste.

2.2.2. Determination of hydration degree by software modeling

The hydration of cements is presumed to take place via dissolution and precipitation processes [19]. To simulate these processes the most well-known and used in many studies [6, 19, 20] and also in this one, is the model proposed by Parrot and Killoh. This model is mathematically described by Equations (15)÷(17), which represent nucleation and growth, diffusion, and formation of a hydration shell, respectively [19].

$$R_{t1} = \frac{K_1}{N_1}(1-a_t)\big(-ln(1-a_t)\big)^{(1-N_1)} \tag{15}$$

$$R_{t2} = \frac{K_2(1-a_t)^{2/3}}{1-(1-a_t)^{1/3}} \tag{16}$$

$$R_{t3} = K_3(1-a_t)^{N_3} \tag{17}$$

where:

R_t- is the controlling rate from the above equations, considered the lowest value of hydration rate for any time step; *K*, *N* and *H* are the associated empirical parameters with the values proposed by Lothenbach et al. [19];

a_t- is the hydration degree of clinker mineral at the time *t*, calculated from the hydration degree of the mineral at the previous time step (a_{t-1}), the time interval (Δ_t), and hydration rate of the clinker mineral at the previous time step ($R_{t1,2,3-1}$) [6, 19, 20]:

$$a_t = a_{t-1} + \Delta_t \cdot min(R_{t1,2,3-1}) f_{w/c} \beta_H \frac{A^{\left(\frac{E_a}{R}(\frac{1}{T_0}-\frac{1}{T})\right)}}{A_0} \tag{18}$$

where:

$$f\left(\frac{w}{c}\right) = 1;\ pentru\ a_t \leq H * w/c \tag{19}$$

$$f\left(\frac{w}{c}\right) = 1;\ pentru\ a_t \leq H * w/c \tag{20}$$

$$\beta_H = \left(\frac{RH-0.55}{0.45}\right) \tag{21}$$

where:

w/c- is the water to cement ratio;

A- is the Blaine surface area of cement (m^2/kg);

A_0- is the reference surface area of cement (385 m^2/kg);

E_a- is the apparent activation energy of clinker mineral (J/mol);

T_0- is the reference temperature (293.15 K);

RH- is the relative humidity, which is 1 in fully hydrated systems, and H^{m} is the critical degree of the clinker mineral.

3. RESULTS AND DISCUSSION

3.1. HYDRATION TROUGH EXPERIMENTAL METHOD

The contet of chemical bound water, calcite and Portlandite produced during cement hydration was estimated by analyzing the DTA/TGA diagrams obtained from the thermal analyses. Figure 1 shows the DTA/TGA curves evolution of the analized cement pastes during 270 days of hydration.

Tipically to cement paste TGA curve shows three significant weight loss steps. The first step at about 100^{o}C has to do with the drying (capillary pores residual water) and/or with the dehydration of ettringite [16]. The second weight loss step at about 420^{o}C-450^{o}C is due to the dehydroxylation of $Ca(OH)_2$. The third weight loss step at about 700^{o}C can be attributed to the decarbonation of $CaCO_3$.

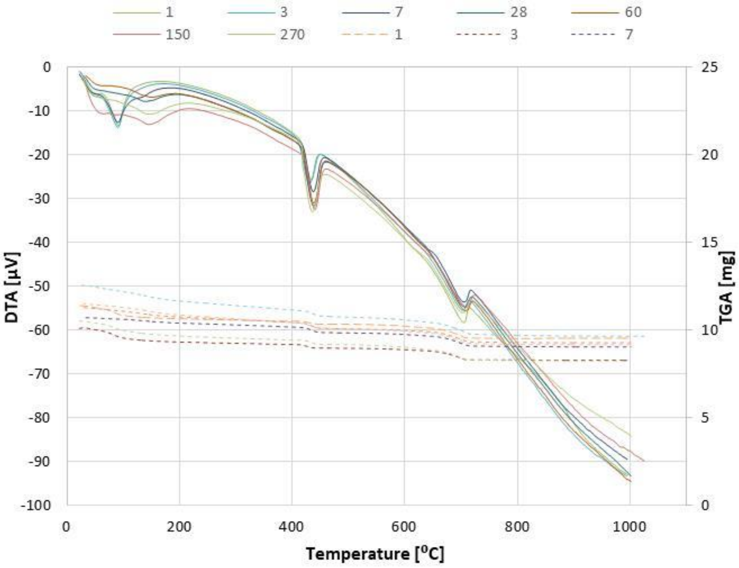

Figure 1. TGA/DTA diagram for a cement paste based on CEM II aged 270 days

The dehydroxylation process of Portlandite has been observed under different atmospheres, for temperatures ranging from 380°C to 550°C [5, 7, 8, 18, 21-23]. In this study, the reference range for Portlandite quantification was 420-540°C. Moreover, when quantifying the total CH content, the possibility that a small part of it has carbonated must be taken into account and corrections must be made in this regard. Carbonated CH content can be calculated based on CO_2 loss from $CaCO_3$ decarbonation according to Wang et al. [18] but it must be keep in mind that the CO_2 can also come from the addition of limestone in the cement. For $CaCO_3$ decarbonisation studies reveal different temperatures of range between 540-850°C [5, 11, 18, 21, 23].

Figure 2 shows the evolution of the content of Portlandite and calcite during 270 days of hydration. It can be seen that the Portlandite amount grows with the hydration period while the calcite amount has small variations around the value of the concentration in which it was added, most likely generated by the occurrence of the Portlandite carbonation process.

In the estimation of the amounts of the Portlandite and calcite the smallest contribution of the carbonation process was took into account and the proper corrections were made. The values that were obtained are similar to those obtained by Soja et al. [11] for a cement paste made from a similar cement powder before exposing the cement paste to CO_2.

Regarding the decomposition of chemically bound water, according to the specialized literature [5, 24, 25] it occurs normally between 105-1000 °C but there are also studies like Panea's [7] that give an interval of 140-1100 °C. In this study, the evolution of chemically bound water content during cement hydration was quantified between 105-1000°C and is shown in Figure 3 where it can be seen that it is directly proportional to the hydration period.

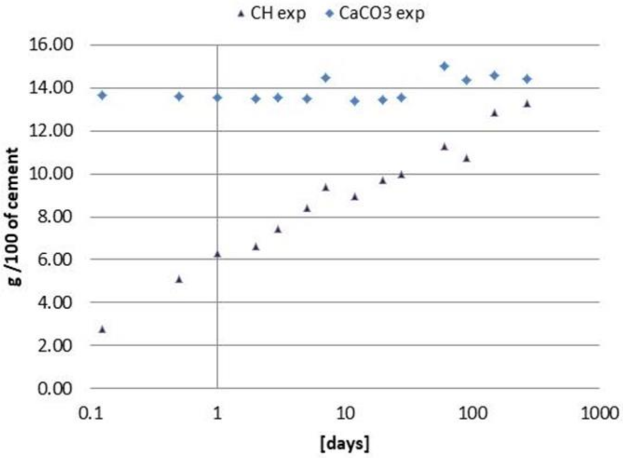

Figure 2. Evolution of the content of portlandite and calcite estimated from DTA/TGA diagrams

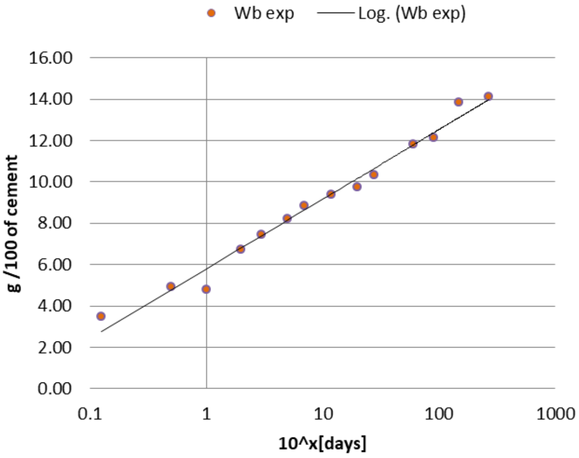

Figure 3. Evolution of chemically bound water estimated from DTA/TGA diagrams

Table 2 shows the total theoretical amount of Portlandite and chemically bound water obtained using the simplified equations (1)÷(8). Based on these theoretical values and the DTA/TGA diagrams the equations (9), (12) and (13) were solved thus obtaining the hydration degree of cement paste presented in

Table 3. It can be observed that the degree of hydration obtained with Method 1 and 3 is almost the same; this was to be expected since the methods are based on the same considerations (Portlandite quantification).

Method 2, which uses chemically bound water to estimate the degree of hydration, shows a much lower degree of hydration of the cement paste. The accuracy of this method can be affected by the carbonation process and a too-high LOI value according to Chu et al. [5].

Table 2. Theoretical amount of CH and bound water after complete hydration of CEM II cement

Cement phase	Mass content in cement [%]	CH mass [g/g of cement]	W_b [g/g of phase]	W_b mass at complete hydration [g]
C_3S	22.085	0.093	0.24	5.300
C_2S	39.843	0.051	0.21	8.367
C_3A	8.625	0	0.40	3.450
C_4AF	9.538	0.014	0.37	3.529
Ca_{free}	0.461	0	0.33	0.152
Gypsum	4.308	0	0	0
Total	-	0.159	-	20.798

Table 3. The degree of hydration of the cement paste using the three experimental methods

Method	Degree of hydration [%]
I	82.080
II	67.840
III	83.528

3.2. HYDRATION THROUGH SOFTWARE MODELING

In this study, the model proposed by Parrot and Killoh for cement hydration was used in GEMS and PHREEQC to estimate the dissolution and hydration degree of all cement clinker phases (C_3S, C_2S, C_3A, C_4AF) as a function of time. The results are presented in Fig. 4 and show mostly the same dissolution kinetics of the clinker phases with a slight degree of dissolution in the case of the PREEQC model for the C_2S phase.

The hydration process starts with the rapid dissolution of C3A known for its reactivity and in the absence of gypsum, the formation of this phase can lead to an undesirable instantaneous setting. C_3S, the most important phase for the mortar strength development during the first 28 days of hydration, dissolves congruently and quite rapidly after coming in contact with water. C2S and C4AF react much slower and contribute to the long-term strength of the cement-based mortars [26].

The dissolution of C_3S and C_2S adds silicate ions to the solution which leads to growth of calcium silicate hydrate gel (C–S–H) and Portlandite. Figure 5 shows how the GEMS software predicts the evolution of Portlandite concerning the hydration period. An important thing that can be observed is that the amount of Portlandite obtained by software modeling is similar to that obtained theoretically.

Dissolution of C_3A and C_4AF adds aluminate ions to the solution, needed for the growth of minerals such as ettringite or calcium monosulfoaluminate [27]. Their appearance and formation are not the subject of this study and will be discussed in another research.

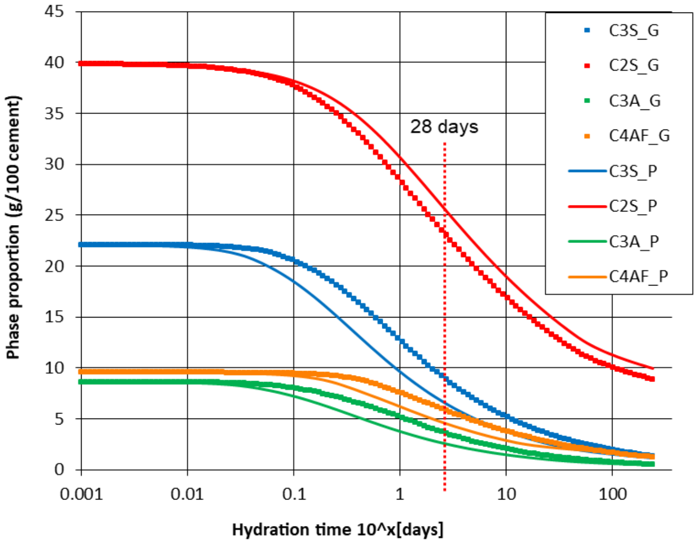

Figure 4. Dissolution kinetics of main cement phases. PHREEQC (P extension) outputs and GEMS outputs (G extension)

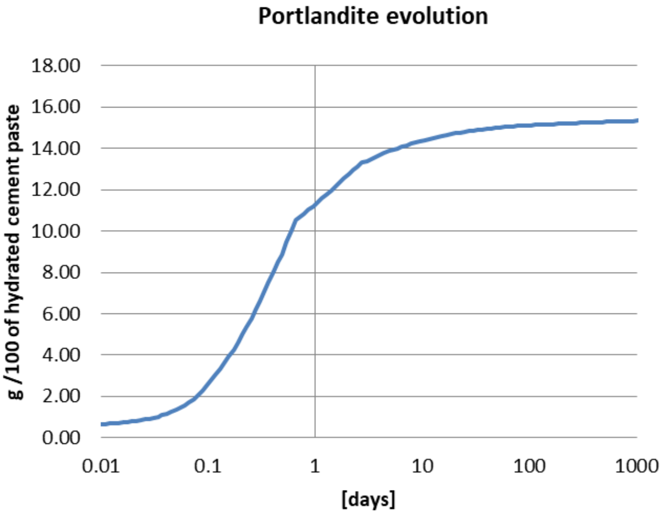

Figure 5. Portlandite evolution during the hydration process of the cement paste

3.3. COMPARISON BETWEEN EXPERIMENTAL AND MODELING APPROACH

A comparative study was performed between data resulting from software modeling and data resulting from the experimental methods. The results are presented in Figs. 6 and 7.

In Fig. 6 it can be seen that in the case of Portlandite, the evolution obtained by modeling has a faster increase compared to that obtained by the experimental method but both tend to the same value as the hydration period approaches 270 days. Regarding calcite, it can be observed that the two models diverge slightly towards the end of the hydration period due to the occurrence of the CH carbonation process, which leads to an increase in the amount of experimental calcite.

Figure 7 shows that the hydration kinetic has the same trend for both experimental and modeling approaches starting with some greater difference in the early stage of hydration. Moreover, there are studies like that of Panea and Hansen [7] which attests that DTA analyses combined with TGA analyses are more suitable for studying hydration at later stages.

Regarding the experimental methods used, in Table 4 it can be seen that the coefficient of determination (R^2) that provides the extent of how well data outcomes from experimental methods replicate the software model shows that Method 1 has a better fit than the other two methods. This may be a consequence of the fact that Method 3 doesn't take into account the probability of Portlandite carbonation and Method 2 uses chemically bound water to estimate the degree of hydration, the accuracy of which is affected by the carbonation process and a too high LOI value cement.

Table 4. Measure of how well data outcomes from experimental methods replicate the modeling data

Software model	The coefficient of determination R^2		
	Method 1	Method 2	Method 3
GEMS	0.979	0.890	0.922
PHREEQC	0.955	0.861	0.890

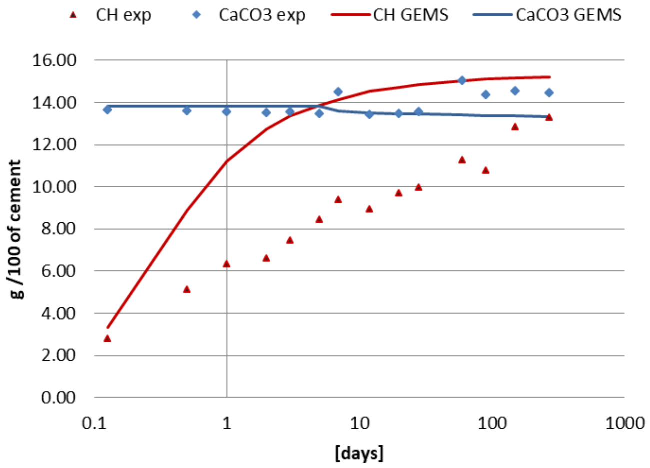

Figure 6. Comparative study of the evolution of Portlandite and calcite amount

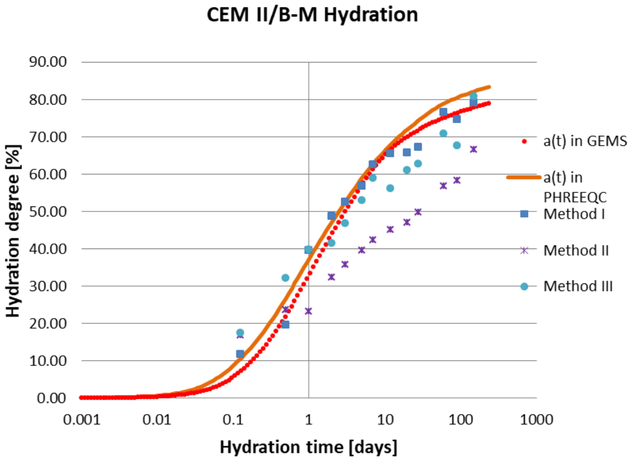

Figure 7. Hydration degree comparative studies between experimental and modeling approach

4. CONCLUSIONS

The results have shown that there is a relatively good agreement between cement hydration kinetic based on portlandite and calcite quantification obtained by thermal analyses (DTA/TGA) and hydration kinetics obtained using GEMS and PHREEQC geochemical codes. As the DTA/TGA analyses are recognized to be suitable for studying the hydration at later stages, the experimental results overestimate the hydration in the early stage of the process. This reinforces not only the idea that modeling can be a powerful tool that can be used in the understanding of reactions and processes occurring in this complex multicomponent-multiphase system of the hydrated cement but also the correctness of the representation of chemical reactions, mathematical models, and parameters used in the two modeling codes.

Understanding these processes and how to program and operate with these modeling codes can facilitate the assessment of the geochemical evolution of cement-based materials under different environmental conditions. This evolution can hardly be reproduced and estimated at the laboratory level, due to the large time scales time involved in the safety assessment of a radioactive waste disposal facility in which cementitious materials are used as engineered barriers.

Currently, it was studied the influence of temperature on the hydration process, and in the future, it intends to develop models that can simulate the chemical degradation of concrete to obtain input information that can be used in predicting the behavior and durability of cement-based materials in various environments, a very important aspect for demonstrating the safety functions associated with cementitious engineered barriers in a radioactive waste repository.

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