

Activation Energy and Heat of Hydration rate for Ceramic Produced in Saudi Arabia, Chinese, Emirates and, Germany

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Abstract: The activation energy E_a 's dependency on the water/solid ratio and the rate of the hydration process is demonstrated by the results obtained using isothermal calorimetry. The activation energy decreases as the water-to-solids ratio increases. The hydration response rate happened during three time periods: the rapid phase, the long-term dormant phase, and the termination phase. At lower hydration temperatures of 25°C and 35°C, the hydration response is strong with a higher water-to-solid ratio. At higher temperatures (60°C and 80°C), the water/solid ratio increased and the density of hydration decreased. Investigated and depicted were the effects of temperature and the water-to-solid ratio on the heat of hydration reaction rate.

Keywords: Ceramics; Heat of hydration; Calorimeter; Activation energy; degree of hydration

1. Introduction

A ceramic is a solid that has been heated and then cooled (Freiman *et al.*, 1991). For example, a ceramic may be either crystalline or amorphous (like glass). For example, a ceramic may be either crystalline or amorphous (like glass). Clay, either on its own or combined with other materials, is the primary ingredient in ceramics and is fired to a hardened state (Iaich *et al.*, 2021). Ceramics are then polished and burned until they develop a soft surface and take on color. Ceramics are used in the construction, industrial, and artistic industries. A new kind of ceramic that can be utilized as a semiconductor material has recently been created. "There are four main categories of ceramics, which are shown below along with some illustrations. The raw materials used to create ceramics do not frequently include clays when they are employed for structural reasons, such as in the production of bricks, pipes, floor tiles, and roof tiles (Ruan *et al.*, 2023). Powder synthesis, powder mixing, thermal treatment, and sintering—the process of spreading the solid state to create the liquid and granular phases—are all steps in the production of ceramic components (Liu *et al.*, 2022). Ceramic tiles are in style and can be created affordably in an array of stunning hues, textures, and patterns. In actuality, the ideal reaction conditions determine the physico-chemical characteristics of tiles. To put it another way, during the design and

manufacturing choosing raw materials with an optimum ratio mixture is very important. Moreover, the impurity of different oxides may result in undesirable colors, while the purity of the raw materials influences the fired products (Ezeh & Onukwuli, 2021). Therefore, two types of kaolinitic clays, namely china clay and ball clay, are widely used as the raw materials for the production of ceramic tiles due to providing a higher degree of plasticity. However, there are other types of raw materials that can be used, including limestone, dolomite, and marble, which are recognized as sources of carbonate. This is important because carbonate acts as a fluxing mineral, which means that its presence may be reduced due to the possibility of it having an unfavorable effect on the size and shape of the formed ceramic (Zhang *et al.*, 2021). In light of the above, the aim of this study was to determine the concurrent impacts of the temperature and the water/solid ratio on the heat evolution of the hydration reaction, the rate of the heat hydration reaction and the associated kinetic parameters, and the activation energy of the hydration reaction.

2. Methodology

2.1 Materials Experimental

2.1 Instruments

The conduction calorimeter used for measuring heat of hydration of ceramics was conduction calorimeter (ZIAC, German Academy of Science) (Smrèková *et al.*, 1996)

2.2 Experiments

2.2 Materials

The materials utilized in this inquiry were gathered from the local market among various varieties of ceramic tiles that are made in Saudi Arabia, the United Arab Emirates, Germany, and China, among other nations. Chemical analysis is used to determine the composition of these ceramics' chemical oxides, as illustrated in Table 1.

Table 1 : The chemical Oxides of Ceramics

Oxides	CaO	MgO	K ₂ O	Na ₂ O	Fe ₂ O ₃	TiO ₂	Al ₂ O ₃	SiO ₂
% wt China	0.1	0.3	1.6	0.1	0.6	0.03	37	48
Saudi Arabia	0.04	0.03	0.61	0.29	0.03	0.10	45.2	23.27
% wt Emirates	0.1	0.1	2.9	0.9	0.3	0.4	23.7	71.6

2.3 Methods of ceramics preparation

2.3.1. The preparation of the ceramic's samples

To a certain fineness and the capacity to pass through a 40-mm sieve, ceramic tiles were milled. With water/solid ratios of 0.5 and 0.7 by weight, four distinct ceramics made with raw materials from various nations were created. Table 2 displays the acronyms related to the samples. Using a conduction calorimeter, measurements of the heat development of ceramic powders at water/solid ratios of 0.5 and 0.7 were made at various temperatures (25°C, 35°C, 60°C, and 80°C). Using rods in a calorimetric cup, the water and ceramic samples were mixed. The closed calorimetric cup was then inserted into the apparatus to enable the monitoring of heat evolution.

2.3.2 The experiment in detail

A conduction calorimeter purchased from the German Academy of Science was used to calculate the heat evolution of each hydration reaction (ZIAC). Two delicate cells were placed inside the

calorimeter's isolating metallic body. The standard sample, which served as a benchmark for the cell, was in the first section, and the hydrating cement sample was in the second. The system was covered in polyuret, a dense foam that served as a barrier between the metallic body and the lid.

Table 2 : Ceramics designated by the following symbols :

CS	CG	CE	CCH
SAUDI CERAMICS	GERMANY CERAMICS	EMIRATES CERAMICS	CHINA CERAMICS

A converter module was used to track the temperature rise in the hydrating cement after the exothermic hydration reaction started by transferring transducer heat between the two cells of the system. Four samples connected to an inert reference sample might be measured by the calorimeter at once. As a result, measurements may be made between the temperatures of 10°C and 90°C. Four measurements of 3 g ceramic samples made in four different nations (Saudi Arabia, the United Arab Emirates, China, and Germany) were made at 25°C, 35°C, 60°C, and 80°C with water/solid ratios of 0.5 and 0.7. Analysis and plotting of the device's data collection. Using a calorimeter, the heat evolution was calculated as a function of temperature at various water/solid ratios. The measurements of the ceramic sample temperatures over time were the result of the experiments. The following equation ([Moutaoukil et al., 2023](#)) determines the formula used to calculate the rate of heat evolution:

$$q_t = \frac{dq}{dt} \quad (1)$$

This equation shows a relationship between the rate of heat evolution and time for this test. Arrhenius equation gives a relation between k rate of reaction with temperature, and it is expressed as follows:

$$k = A.e^{-\frac{E_a}{RT}} \quad (2)$$

Where: k is rate constant temperature, dependent it equals the rate of heat evolution, and it is assumed to affirm the Arrhenius relationship, R equals the natural gas constant (8.314 J/mol/K), T is temperature, A is a temperature-independent constant of proportionality or a proportionality constant (same units as k) in this reaction it is neglected, and E_a (J/mol) is activation energy (J/mol), measurements of reaction rates can be made at different isothermal temperatures ([Li et al., 2019](#)). it is obtained experimentally by drawing the log of reaction rate versus the inverse of the reaction temperature $\ln(k)$ versus $1/T$. E_a calculated from the negative slope with independent of A, because the value A is not reported in ceramics hydration research, (A) is therefore neglected out. The induction period shortens as temperatures rise, but as temperatures reach higher levels (between 60°C and 80°C), their effects become less noticeable. Temperature increases have an impact on both the change in hydration rate and the peak's form. When the water/solid ratio is smaller, the rate of dissolution is affected by the water content, which aids in the formation of the principal hydration products all over the ceramic grains. The sensitivity of the Arrhenius theory to temperature hydration It is the best equation to determine how temperature affects the rate of hydration and is appropriate to explain all chemical processes ([Li et al., 2022](#); [Kulsartov et al., 2021](#)). E_a calculations ought to be accurate enough to distinguish between samples of various ceramic materials and compositions.

3. Results and Discussion

3.1 Scanning Electron Microscope (SEM)

Ceramics pastes samples were fixed on a glass plate with petroleum wax, and a thin film of gold was sprayed for SEM analysis. In the vacuum chamber of the gold depositing machine, gold particles were deposited on the surface of the palm frond. Special conductive liquids were used to make the glass plate conductive as well. [Figure 1](#) shows scanning electron micrographs of cement surfaces before they were burned and converted to ash. The results revealed that the surface of the ceramic pastes was rough and undulating. After calcination, the morphology of cement paste was significantly destroyed, and asperities on its surface were almost completely removed.

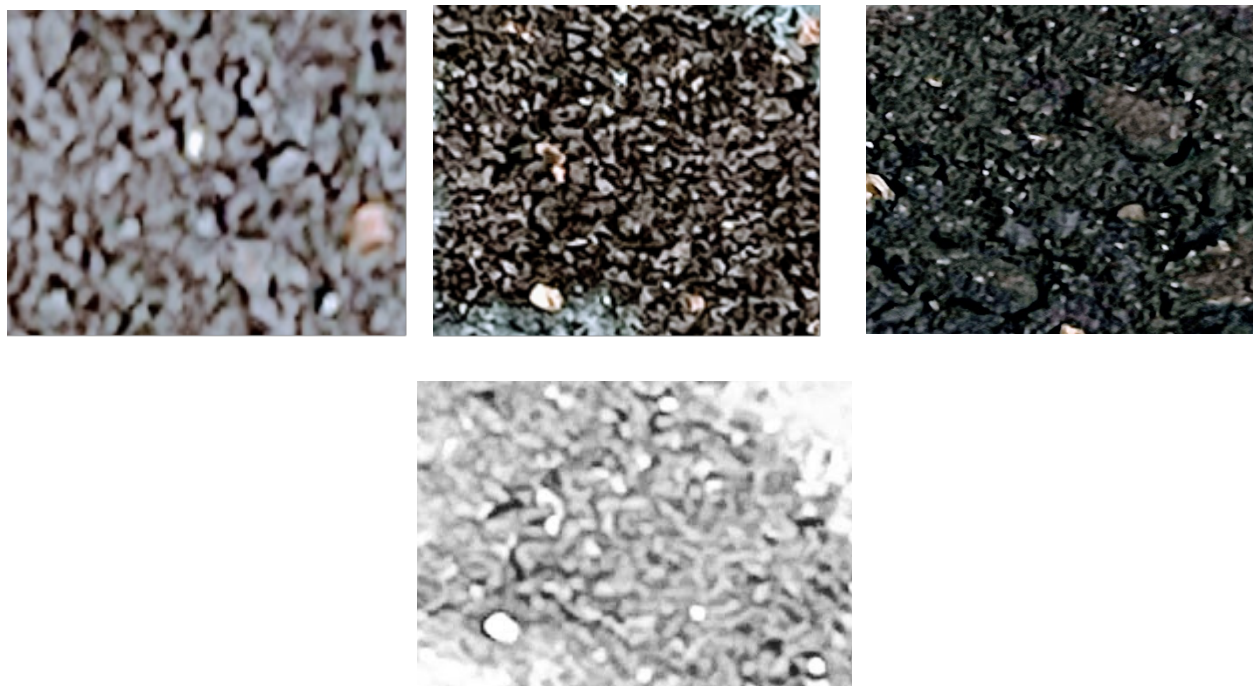


Figure 1: SEM images of ceramics pastes

3.2 Influence of water/solid ratio and Temperature on heat of hydration

The calorimetric curves, which are depicted in [Figures 1-6](#), demonstrate the direction in which heat evolution takes place during the hydration reaction. These graphs describe how temperature affects the calorimetric curves at various water-to-solid ratios. The following periods can be used to divide up the processes in the hydration process. When water was added to the ceramic powder, a quick reaction step occurred, and heat was shown to develop from it (as indicated by a little peak in [Figures](#) (2–7)). The calorimetric curves, which are depicted in [Figures 1–6](#), demonstrate the direction in which heat evolution takes place during the hydration reaction. These graphs describe how temperature affects the calorimetric curves at various water-to-solid ratios. The following periods can be used to divide up the processes in the hydration process: When water was added to the ceramic powder, a quick reaction step occurred, and heat was shown to develop from it (as indicated by a little peak in [Figures 2–7](#)). the magnitude, duration, and primary hydration peak. The figures illustrate how temperature and the water/solid ratio affect the kinetics and process of hydration. The increased temperature has an impact on the brief induction period. When the temperature rises, the form of the primary peak changes due to the altered hydration mechanism, making the thermal influence at higher temperatures (60°C and 80°C) on the extent of the induction insignificant and hence negligible. At a lower water/solid ratio,

first hydration products are formed around ceramic samples on the grains, where the dissolving rate is dependent on the water content. These grains act as a barrier that prevents the diffusion of ions and water. It's plausible that temperature influences, Hence, based on this supposition, it may be possible to explore whether the through-solution mechanism is responsible for hydration reactions at higher temperatures with excess water compared to lower temperatures with a lower water/solid ratio. Rapid precipitation and a dense hydration rate may occur when the volume of the solution is saturated or supersaturated with ions associated to it. and intense hydration is occurring. By using a through-solution technique and a shorter induction duration, the peak of hydration is shifted.

3.3 Influence of water/solid ratio hydration

Figures 2-7: The potency of hydration heat increases somewhat with increasing water/solid ratios at lower temperatures (25°C and 35°C), demonstrating the influence of the water/solid ratio. In relation to how long the induction period lasted, the hydration happened quickly. Over time, the curves lengthen. The hydration heat peaks are higher and narrower with shorter induction times as the temperatures rise to 60°C and 80°C.

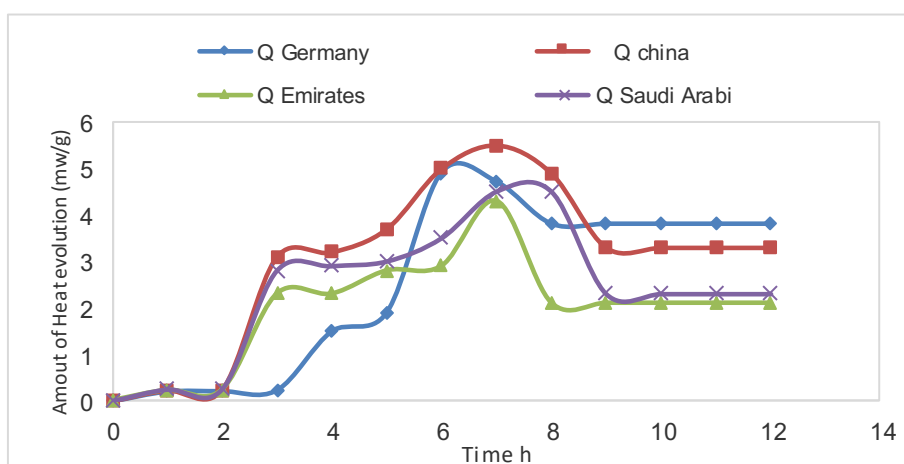


Figure 2. Influence of water/solid (0.5) and temperatures upon hydration heat evolution

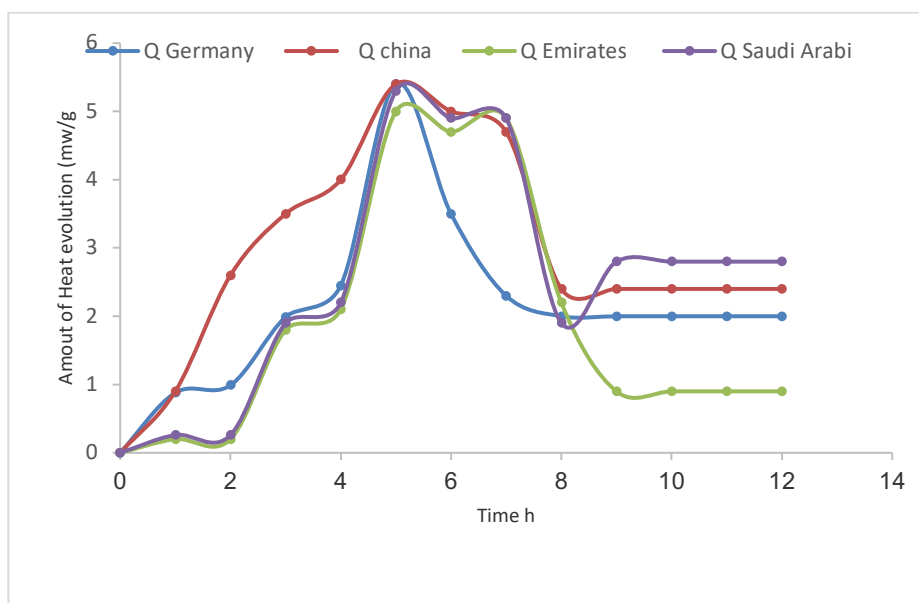


Figure 3. Influence of water/solid (0.7) and temperatures upon hydration heat evolution

In addition to being substantially temperature-dependent, the results of the rate of hydration at 60°C and 80°C under specific water/solid ratios suggest that hydration happens through another method. Low temperatures and a higher water-to-solids ratio favor the surface chemical process. Yet, the via solution mechanism drives the hydration reactions at higher temperatures with elevated water. At lower temperatures (25°C and 35°C), the intensity of hydration heat gradually increases with increasing water/solid ratios. Moreover, the duration of the induction phase is correlated with the acceleration of hydration. Figures 1-6 show how the water-to-solid ratio affects heat production in the calorimeter at lower temperatures (25°C and 35°C), which ends after 3 hours at 80°C and 4 hours at 60°C. When temperatures are higher (between 60°C and 80°C), some water partially evaporates.

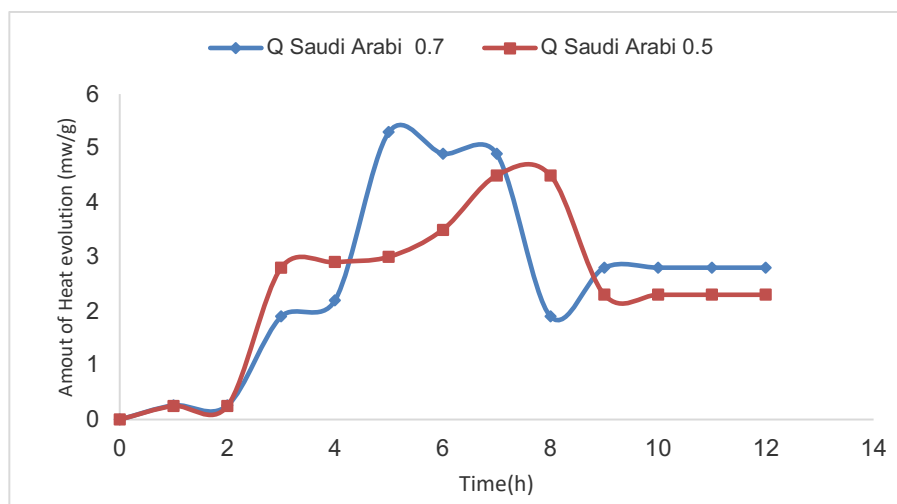


Figure 4.: Influence of water/solid ratio and upon hydration heat evolution 0.5 and 0.7 ceramics, Saudi Arabia

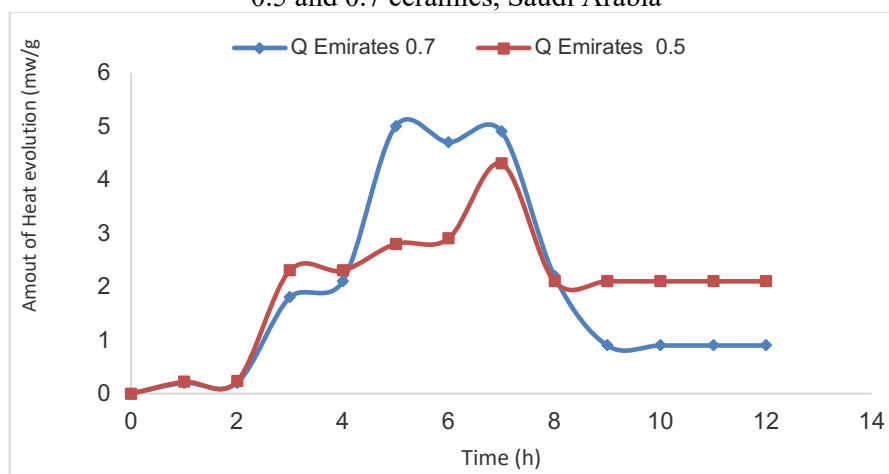


Figure 5. Influence of water/solid ratio and temperature upon hydration heat evolution 0.5 and 0.7 ceramics Emirates

3.3 Determine of activation energy

For all test temperatures T and all test samples, the calculation of E_a is performed at any hydration reaction rate. Four of the mixes evaluated in this study underwent distinct hydration reactions, and the value E_a was calculated for each one. Results for these combinations at water/solid ratios of 0.5 and 0.7 are displayed in Figs. 8 and 9 and in Table 3. Figures 8 and 9 demonstrate how strongly the hydration response affects E_a . In comparing the heat evolution data with the activation energy estimations at different hydration reactions for the test samples, a number of useful conclusions may be drawn.

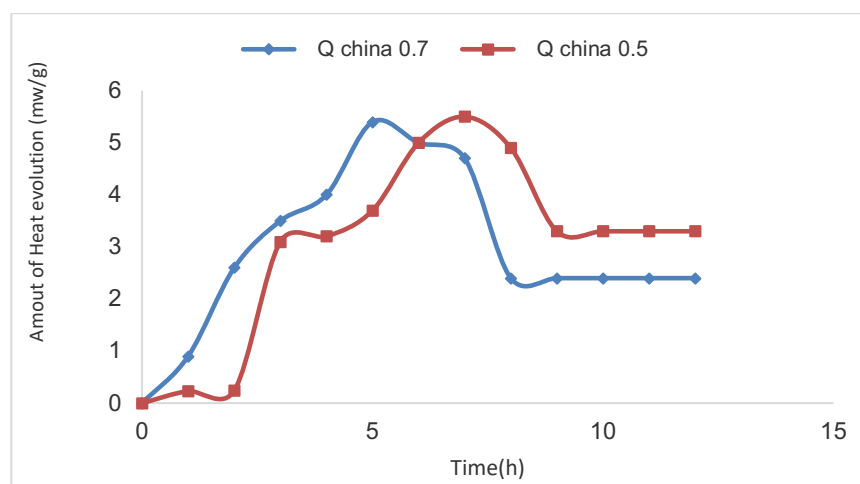


Figure 6: Influence of water/solid ratio and temperature upon hydration heat evolution 0.5 and 0.7 ceramics China

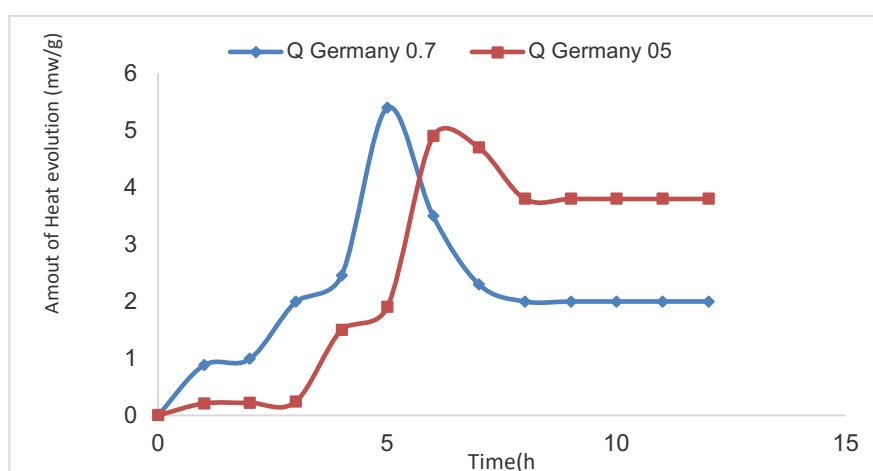


Figure 7: Influence of water/solid ratio and temperature upon hydration heat evolution 0.5 and 0.7 ceramics Germany

Table 3's entries for the data from figures 8 and 9 indicate that the levels of activation energy depend on the water-to-solids ratio. The activation energy decreases as the water/solid ratio increases (Kulsartov et al., 2021); it also depends on the degree of hydration (Yuan et al., 2022). For each test sample, the values of R2 range from 0.92 to 0.99 for the water/solid ratio at 0.5 and from 0.94 to 0.99 for the water/solid ratio at 0.7, indicating accurate characterization of the hydration process.

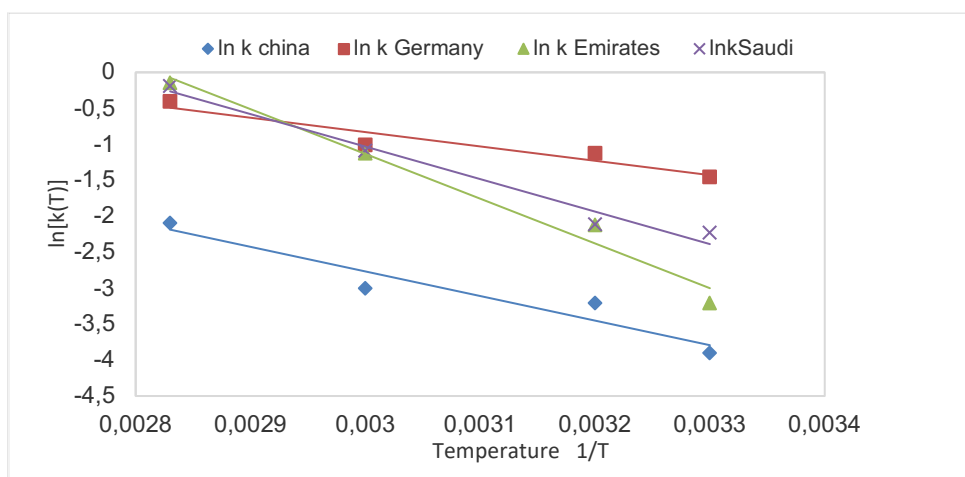


Figure 8. Determination of k using linear fit for the ceramic samples and E_a from plot of $\ln(k)$ versus $1/T$ at 0.5

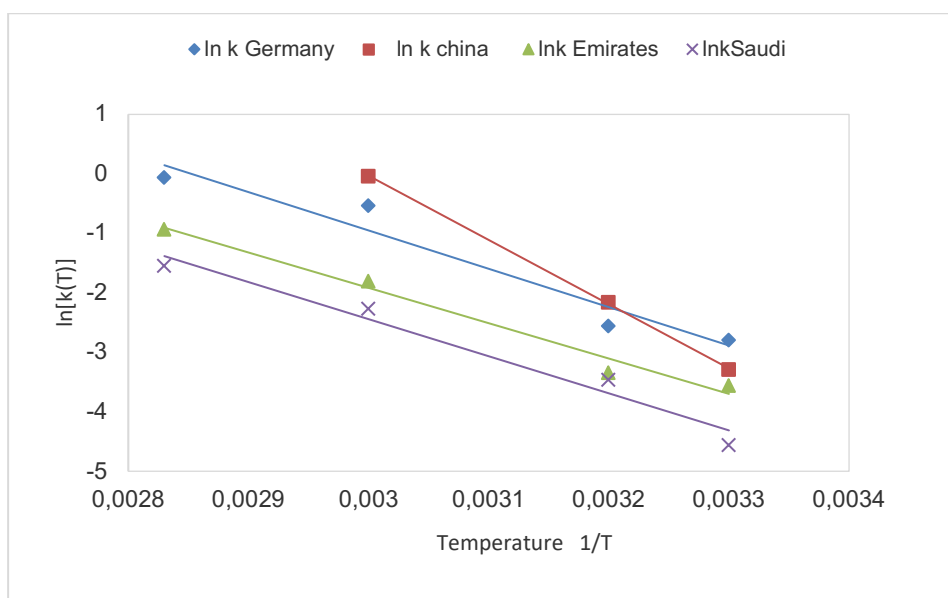


Figure 9. Determination of k using linear fit for the ceramic samples and E_a from plot of $\ln(k)$ versus $1/T$ at 0.7.

Table 3: Activation energy E_a at different water/solid ratio

Types of ceramic	E_a J/mol water/solid ratio 0.5	R^2 water/solid ratio 0.5	E_a J/mol water/solid ratio 0.7	R^2 water/solid ratio 0.7
Saudi	51.89	0.96	20.59	0.96
Emirate	49.41	0.97	21.99	0.98
China	53.66	0.92	22.53	0.99
Germany	53.61	0.91	22.59	0.94

Conclusions

The focus of the current essay is on how temperature and the water-to-solids ratio affect the heat of hydration and the activation energy of ceramics. The resulting data allows for the following inferences to be made: An increase in temperature at a water/solid ratio of 0.5 can speed up the hydration reactions of several types of ceramic powders. At the lower hydration temperatures of 25°C and 35°C, the hydration reaction rate is high and the water/solid ratio is higher. By raising the water/solid ratio to 0.7 at higher temperatures (60°C and 80°C), the density of the hydration can be decreased. The major hydration peak's key characteristic is its appearance with rising temperatures, which denotes a change in the hydration mechanism.

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References

- Copeland L. E.; Kantro D. L.; Verbeck G., 1960 "Part IV-3 Chemistry of Hydration of Portland Cement," *4th International Symposium of the Chemistry of Cement, Washington, D.C.*, pp. 429-465.
- Cui M., Li, X., Wang J., Wang Z., Li L., & Lyu X. (2022). Alkali-hydrothermal activation of mine tailings to prepare one-part geopolymer: Activation mechanism, workability, strength, and hydration reaction. *Ceramics International*, 48(20), 30407-30417

- Ezeh E. M. and Onukwuli O. D. (2021). Comparative Cone calorimetric analysis of the fire retardant properties of natural and synthetic additives in banana peduncle fibre reinforced polyester composites, *Mor. J. Chem.* 9, 530-541, <https://doi.org/10.48317/IMIST.PRSM/morjchem-v9i3.21954>
- Iaich S., Miyah Y., Messaoudi L. (2021). Elaboration and characterization of low cost tubular ceramic supports made of Moroccan clay for microfiltration and ultrafiltration membranes, *Mor. J. Chem.* 9N°2(2021) 185-197
- Kulsartov T., Kenzhina I., Chikhray Y., Zaurbekova Z., Kenzhin, Y., Aitkulov, M., ... & Dyussambayev, D. (2021). Determination of the activation energy of tritium diffusion in ceramic breeders by reactor power variation. *Fusion Engineering and Design*, 172, 112783
- Liu Q., Cui M., Li X., Wang J., Wang Z., Li L., & Lyu X. (2022). Alkali-hydrothermal activation of mine tailings to prepare one-part geopolymer: Activation mechanism, workability, strength, and hydration reaction. *Ceramics International*, 48(20), 30407-30417.
- Li J., & Huang J. (2022). Thermal Debinding Kinetics of Gelcast Ceramic Parts via a Modified Independent Parallel Reaction Model in Comparison with the Multiple Normally Distributed Activation Energy Model. *ACS omega*, 7(23), 20219-20228
- Moutaoukil G., Alehyen S., Sobrados, Fadil M. , Taibi M. (2023)" Effects of temperature, time and alkaline solution content on the mechanical properties of Class C fly ash-based geopolymer using Taguchi method", *Mor. J. Chem.*, 11, Issue 01, Page 61-76
- Ruan W., Liao J., Gu X., Mo J., Cai M., Guo, W., ... & Ma, X. (2023). Effects of bauxite tailings and sodium silicate on mechanical properties and hydration mechanism of magnesium phosphate cement. *Construction and Building Materials*, 366, 130055
- Smrčková E., Palou M.T. & Tomková V. (1996). Application of conduction calorimetry for study of the reactivity of C2S in the system C2S-C4A3⁻S-C⁻S-H. *Journal of Thermal Analysis*, 46, 597–605 <https://doi.org/10.1007/BF02135039>
- Yuan W., Kuang J., Huang Z., & Yu M. (2022). Effect of aluminum source on the kinetics and mechanism of mullite preparation from kaolinite. *Chemical Physics Letters*, 787, 139242.
- Zhang J., Pan G., & Yan Y. (2021). Early hydration, mechanical strength and drying shrinkage of low-carbon alkali-activated Ti-extracted residues-fly ash cement and mortars. *Construction and Building Materials*, 293, 123517.

(2023) ; <https://revues.imist.ma/index.php/morjchem/index>