Calcination process and kinetic carbonation effect on the hydrated and anhydrate phases of the OPC matrix at early age of hydration

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ABSTRACT

This study aims to improve the understanding of the effect of both calcination temperature and the carbonation process on cement pastes by analyzing the Fourier-transform infrared spectroscopy (FTIR) principal vibration modes. In this regard, six samples of cement pastes, identified as (CS1, CS2, CS3, CS4, CS5 and CS6), mixed with white sugar with different concentrations ($X = 0$, 0.2, 0.4, 0.6, 0.8, and 1.1 \%), respectively, were prepared. These blended cement samples were heated (calcined) at a temperature of about 600$^\circ$C. Moreover, an additional sugar-free sample tagged as CS0 was prepared and cured at room temperature for 7 days. Afterward, these samples were subjected to FTIR spectroscopy and X-ray diffraction to investigate the effect of the calcination and the carbonation process on the principal mode of vibrations and the microstructure of the ordinary Portland cement (OPC) matrix as well. The characteristics of the FTIR principal modes of vibrations were investigated. Besides, the relaxation time and the rotational energy barrier were calculated for different sugar concentrations. This study revealed that the kinetic reaction of the carbon contained in the different samples with the C\textsubscript{3}S, C\textsubscript{2}S, C–S–H and CH phases leads to the formation of calcium carbonate CaCO\textsubscript{3}, where the rate of formation depends upon the carbonation rate of samples. The critical variation for the relaxation time and rotational energy barrier clearly appeared at $X = 0.6\%$. Moreover, the kinetic carbonation of both clinker phases and the hydrated phases in the OPC matrix system mainly depends upon the rate of the polymerization of the silicate group inside the matrix of OPC.

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**Introduction**

The ordinary Portland cement (OPC) paste is considered as the principal component of mortar and concrete. Therefore, the hydration of cement paste is of greatest importance in making high-quality concrete products [1–5]. Cement hydration is a chemical process that forms the anhydrous material through several chemical reactions, leading to the formation of hydrates. Among the different factors that affect the rate of cement hydration comes the curing temperature. It was shown that the rate of hydration increases by the curing temperature [6–10]. It should be noted that the available experimental results regarding the effects of elevating the curing temperature on hydration kinetics are still limited. There are few publications that consider curing temperatures higher than 60°C (333 K) [11].

The thermal treatment (calcination) of the blended cement paste at a temperature (600°C) leads to an improvement in the decomposition of the hydration products and in the kinetic carbonation reaction with the cement matrix system’s chemical phases. The chemical process that is triggered by the effect of calcination temperature and carbonation reaction of cement matrix has an important influence on the physical and chemical deformation inside the cement pastes. This effect leads to an increase in decomposition of the hydration products effecting the principal anhydrite phases of cement material. The deformation in the physical and chemical properties occurs due to the formation of calcium carbonate (CaO\(_2\)) and the appearance of some microcracks in the structure cement matrix. These changes lead to a destruction of the building force inside the cement matrix. The carbonation process and the calcination temperature effect can be considered as a hazard process on the durability of the cementitious materials due to the chemical reaction of carbon dioxide with both the clinker and hydrated phases of the cement matrix. This process induces the decreasing of the reinforcing of the cement materials, causing severe damages to the building structure [12–18].

This article aims to study the effect of both calcination temperature and carbonation process in cement pastes by analyzing the FTIR principal vibration modes. In order to achieve this, cement samples were blended with different concentrations of table white sugar, which acts as an artificial source of carbon dioxide through calcination process. The samples were subjected to X-ray diffraction (XRD) to investigate the effect of the calcination and the carbonation process on the microstructure of the OPC matrix.

**Experimental materials**

The used cement was the OPC, with the chemical composition shown in Table 1.
Table 1: Chemical analysis of the used OPC samples.

|   | SiO₂ | TiO₂ | Al₂O₃ | Fe₂O | FeO  | MgO | CaO | Na₂O | K₂O | P₂O₅ | H₂O | SO₃ | Cl |
|---|------|------|-------|------|------|-----|-----|------|-----|------|-----|-----|----|
| OPC | 20.85 | 0.28 | 4.7   | 3.86 | 0.16 | 1.6 | 61.71 | 0.967 | 0.25 | 0.61 | 0.68 | 2.9 | - |

Table white sugar is the common name for a sugar known as sucrose. It is a type of disaccharide made from the combination of the monosaccharide glucose and fructose. The chemical or molecular formula for sucrose is C₁₂H₂₂O₁₁.

**Experimental methodology**

**Preparation of samples**
Six sugar-blended cement samples tagged as CS1, CS2, CS3, CS4, CS5, and CS6 were prepared by mixing 2 g of OPC of each sample with different concentration of sugar 0, 0.2, 0.4, 0.6, 0.8, and 1.1%, respectively, on cement wt% basis, together with a suitable amount of clean distilled fresh water free from impurities and organic substances. The water to cement ratio (W/C) for all samples was kept constant at a value of 0.25. The blended cement samples were heated at a temperature of 600°C and were then cooled to room temperature ≈ 27°C. After that the different samples were ground into a fine powder. Moreover, an additional sample tagged as CS0 free of sugar was prepared and cured at room temperature for 7 days. Thus, it can be shown that both samples CS0 and CS1 are sugar-free; however, CS1 was heated at a temperature of 600°C.

**FTIR measurements**
The FTIR measurements were carried out using a Beckman G.R-7224 spectroscopy meter. The infrared spectrum was recorded in the region 400–4000 cm⁻¹ on graphical paper with a scanning time of 30 min.

**XRD measurements**
XRD measurements were carried out by using X-ray diffractometer Shimatzu-6000, with a scan rate 2° per minute from 5° to 80° (2-theta), with Cu-Kα radiation generated at 25 mA and 40 kV. The quantitative phase analysis was performed using Match3 software.

**Results and discussion**
The phase decomposition of both hydrated and dehydrated phases of OPC matrix plays an important role in the performance (i.e. stability and durability) of any cement-based materials. So, it was necessary to study the physical processes that would affect the formation of both the hydrated products and
the clinker phases. Studying the effect of calcination process and the carbonation reactions with the hydrated phases leads to a better understanding for physical and the microstructural properties of the OPC system matrix. In this respect, FTIR spectroscopy and XRD techniques are employed. This section includes detailed analyses for both the FTIR principal modes of vibrations and XRD spectra for the prepared samples with different sugar concentration.

**Quantitative analysis for the FTIR measurements**

**FTIR spectra**

The chemical process triggered by elevated temperature in the particular phases of cement has a significant impact on the deformation of the physical features, such as the FTIR principal modes of vibrations, of the cement matrix system. The FTIR spectra of both the sugar-free samples CS0 and CS1 are shown in Figure 1, where the influence of the calcination temperature on the FTIR principal modes of vibration for the different cement hydrated phases calcium silicate hydrate (CSH), calcium hydroxide (CH), and the clinker phases tri-calcium silicate ($C_3S$) and di-calcium silicate ($C_2S$) is illustrated.

The FTIR principal modes of vibration for both sugar-free samples CS0 and CS1 are summarized and listed in Table 2. It can be seen that there are clear variations in the principal modes of vibration. These variations appear in the relative intensity (RI), transmittance, and band shape, as well as in the

![Figure 1](image_url). The effect of calcination on the FTIR spectra of OPC: at room temperature (CS0) and calcination temperature (CS1).
frequency shift of the vibrational modes. These variations are observed in the silicate group (ν₄) (Si–O) (460–524 cm⁻¹), the out-of-plane bending mode (ν₂) (CO₃⁻) (842–910 cm⁻¹), stretching S–O mode (ν₃) (SO₄²⁻) (1100–1200 cm⁻¹), the asymmetric stretching mode of (CO₃⁻)(ν₃) (1420–1590 cm⁻¹), and the symmetric and antisymmetric stretching mode of O–H group (3640–3790 cm⁻¹) [16–19]. It can be seen that the latter O–H vibrational frequency band (3640–3790 cm⁻¹) that appears in the case of the naturally hydrated sample CS0 does not appear completely for the calcined sample CS1. This can be attributed to the depletion of CH hydrated phase due to the reaction with the CO₂. Besides, it can be seen that the silicate group (ν₄) (Si–O) (460–524 cm⁻¹) that is responsible for the production of the CSH hydrated phase still exists. Moreover, the decrease in the RI in the symmetric and the antisymmetric stretching of (O–H) group (3100–3500 cm⁻¹) is attributed to the increase in loss of bound water after calcination, which leads, in turn, to an increase in both the pore system and the activity of the carbonation reactions with the OPC matrix. The effect of calcination temperature (600°C) on the hydrated (early hydration) cement matrix may be due to the formation of ettringite and calcium aluminate hydrates (C₆AH₃) due to the reaction of calcium aluminate (C₂A) with the leaching of Ca⁺⁺ from the liberated lime during the hydration of OPC system and the decomposition of the CH hydrated phase, and the loss of bound water due to the CSH hydrated phase decomposition [20–22].

The kinetic carbonation reaction possesses a greater risk as it causes the disintegration of the binding matrix. The carbonation of cement-based materials is a complex mechanism that depends upon the reactivity of the target cement material and carbon diffusion inside the cement matrix. This mechanism is greatly affected, however, by many factors such as the calcination temperature, the carbon concentration, and the degree of hydration. The
calcination process at high temperature is expected to have an important impact on physical properties and the microstructure of the cement-based material.

The FTIR spectra of cement samples CS1, CS2, CS3, CS4, CS5, and CS6, after calcination at a temperature of 600°C, are illustrated in Figure 2. In this figure, the effect of increasing sugar concentration (0 ≤ X ≤ 1.1 %), which implies an increase in the kinetic carbonation rate, on the FTIR principal modes of vibrations is illustrated. It can be seen that the variation in the symmetric and antisymmetric stretching modes of (O–H) group (3100–3500 cm⁻¹) is due to the loss of bound water from the CSH hydrated phases. The rate of change for this band can be considered as an indication for the formation of calcium carbonate CaCO₃ (amorphous state) and the silica gel [23]. The asymmetric stretching mode of CO₃⁻ (1420–1590 cm⁻¹), on one hand, decreases with kinetic carbonation rate up to (X = 0.6%). This is mainly attributed to the decrease of total porosity of OPC matrix due to the precipitation the CaCO₃ inside the pores of this system. On the other hand, it increases for (X > 0.6 %) due to the increase in the relative proportion of the capillary porosity, which leads in turn to an increase in the ionic diffusivity of the carbonation reaction in OPC matrix [24]. The stretching mode S–O (1100–1200 cm⁻¹) (v₃ SO₄⁻) shows similar behavior to that of the previous mode (1420–1590 cm⁻¹) and that is mainly related to the ettringite and gypsum formation due to the

![Figure 2](image-url). The effect of carbonation rate on the different FTIR modes of vibration.
effect of the reaction of the liberated \( \text{Ca}^{++} \) ions with the \( \text{SO}_4^{--} \) and \( \text{CO}_3^{--} \) groups [25]. The out-of-plane bending mode of vibration of \( \text{CO}_3^{--} \) (842–910 cm\(^{-1}\)) has the same trend as the aforementioned modes of vibrations and has a minimum value at \( X = 0.6\% \) and that is attributed to the formation of \( \text{CaCO}_3 \) (calcite). This mode is affected by the total porosity and the relative capillary porosity of the OPC matrix during the carbonation process [24]. The silicate group band mode \( \nu_4 \) (Si–O) (460–524 cm\(^{-1}\)) shows the same trend in all FTIR spectra. In this band, the increase in the carbonation process of the CSH phase leads to an increase in the formation of the silica gel and transform OPC matrix to amorphous structure. Moreover, it shows lower transmittance at \( X = 0.6\% \) due to the minimum formation of gypsum \( \text{CaSO}_4 \). The intensity of the asymmetric stretching band mode (Si–O) (970–1025 cm\(^{-1}\)) has a minimum value at \( X = 0.6\% \), then it increases due to the carbonation reaction of cement clinker phases of \( \text{C}_3\text{S} \) and \( \text{C}_2\text{S} \) (\( \approx 980 \text{ cm}^{-1} \)), which leads in turn to the formation of the calcite (\( \text{CaCO}_3 \)). All of these FTIR modes of the calcined samples are listed in Table 3.

It is now clear that the rate of carbonation of OPC matrix system depends primarily on the silicate groups of various chemical states in the carbonation process, which is different than the standard silica gel, where the kinetic carbonation of the anhydrite clinker phases (\( \text{C}_3\text{S} \)), and \( \beta\text{-C}_2\text{S} \) of the matrix is slower than that of the hydrated phases (CSH and CH) [26]. Accordingly, all the hydration produced phases CSH, CH, and the clinker phases \( \text{C}_3\text{S} \) and \( \text{C}_2\text{S} \) are more affective in the kinetic carbonation process through the following reactions:

\[
\text{C}_3\text{Scarbonationreaction} : 3\text{CaO} \cdot \text{SiO}_2(s) + 3\text{CO}_2(aq) + n\text{H}_2\text{O} \xrightarrow{rC_3S} \text{SiO}_2.n\text{H}_2\text{O} + 3\text{CaCO}_3 \tag{1}
\]

\[
\text{C}_2\text{Scarbonationreaction} : 2\text{CaO} \cdot \text{SiO}_2(s) + 2\text{CO}_2(aq) + n\text{H}_2\text{O} \xrightarrow{rC_2S} \text{SiO}_2.n\text{H}_2\text{O} + 2\text{CaCO}_3 \tag{2}
\]

\[
\text{C – S – Hcarbonationreaction} : 3\text{CaO} \cdot 2\text{SiO}_2.3\text{H}_2\text{O}(s) + 3\text{CO}_2(aq) \xrightarrow{r\text{CSH}} 3\text{CaCO}_3.2\text{SiO}_2.3\text{H}_2\text{O} \tag{3}
\]

\[
\text{CH carbonation reaction} : \text{Ca(OH)}_2(aq) + \text{CO}_2(aq) \xrightarrow{r\text{CH}} \text{CaCO}_3 + \text{H}_2\text{O} \tag{4}
\]

The variations of the RI with the rate of carbonation (i.e. sugar concentration) for both (a) the out-of-plane silicates group (Si–O) (\( \approx 520 \text{ cm}^{-1} \)) mode and (b) the symmetric and antisymmetric stretching of (O–H) group (\( \approx 3435 \text{ cm}^{-1} \)) mode are illustrated in Figure 3a and b, respectively. It can be shown that the variation in the RI depends upon the rate of carbonation (i.e. sugar concentration) of the sample. This result implies that the critical range
| Vibrational band (cm\(^{-1}\)) | Assignment | Compound | CS1 | CS2 | CS3 | CS4 | CS5 | CS6 |
|---------------------------------|------------|----------|-----|-----|-----|-----|-----|-----|
| 460–524                         | \(\nu_4\) – the out-of-plane vibration of Si–O of silicate group | C–S–H hydrated phase | 523.6 | 523.6 | 522.6 | 523.6 | 523.6 | 519.7 |
| 713                             | \(\nu_4\) – out-of-plane bending vibration of CO\(_3^–\) | CaCO\(_3\) calcite | - | - | - | 718.4 | 716.4 | - |
| 842–910                         | \(\nu_2\) – out-of-plane bending vibration of CO\(_3^–\) | CaCO\(_3\) calcite | 868.8 | 878.4 | 883.3 | 877.5 | 915.6 | 878.2 | 872.6 |
| 970 – 1025                      | \(\nu\) – the asymmetric stretching band of Si–O | C–S–H | - | - | - | - | - | 999.9 |
| 1100–1200                       | \(\nu_3\) – stretching S–O bond of SO\(_4^{2–}\) | Ettringite/gypsum | 1144.5 | 1157 | 1158 | 1117.5 | 1154 | 1143.5 |
| 1420–1590                       | \(\nu_3\) – asymmetric stretching of CO\(_3^–\) | CaCO\(_3\) calcite | 1455 | 1433 | 1431 | 1430 | 1424 | 11431 |
| 1600–1700                       | \(\nu_2\) – bending mode of H\(_2\)O | H\(_2\)O | - | 1634 | 1631 | 1629.5 | 1655 | 1644 |
| 1795–1800                       | \(\nu_4 + \nu_1\) of CO\(_3^–\) | CaCO\(_3\) calcite | 1798.5 | - | - | - | 1795.4 | - |
| 2512–2515                       | \(\nu_4 + \nu_3\) of CO\(_3^–\) | CaCO\(_3\) calcite | - | - | - | - | 2512.3 | - |
| 2875–2879                       | - | - | - | 2874.8 | - | - | - |
| 2983 – 2985                     | - | - | - | - | - | - | - |
| 3100–3500                       | Symmetric and anti-symmetric stretching of O–H | H\(_2\)O | 3434.6 | 3435.5 | 3436.5 | 3436.5 | 3435 | 3441 |
| 3640–3645                       | Stretching of O–H of Ca(HO\(_2\)) | Ca(HO\(_2\)) | - | - | - | - | - | - |
for the variation of (RI) for the different samples occurs at $X = 0.6\%$ for both the silicate group and the symmetric and antisymmetric (O–H) group. The variation in the (RI) may be attributed to the differences in polarizability arising from the cationic and anionic ions in the sample occurring at a definite frequency for each principal mode due to the effect of the infrared rays on the samples [27]. This result, which we have reached beforehand, can be consulted to clarify the variations observed in all FTIR spectra shown in Figure 2.

Figure 3. The variation of the relative intensity with the carbonation rate for (a) the out-of-plane silicates group (Si–O) and (b) the symmetric and antisymmetric stretching of (O–H) group.
The variations of the absorbance with the rate of carbonation (i.e. sugar concentration) for both (a) the out-of-plane silicates group (Si–O) (≈ 520 cm\(^{-1}\)) mode and (b) the symmetric and antisymmetric stretching of (O–H) group (≈ 3435 cm\(^{-1}\)) mode are illustrated in Figure 4a and b, respectively. Similarly, it can be shown that the variation in the absorbance depends upon the rate of carbonation (i.e. sugar concentration) of the sample. For the case of the out-of-plane Si–O group, Figure 4a illustrates that the absorbance decreases with the increase of the carbonation rate and reaches an absolute minimum value at \(X = 0.6\%\), and then increases with the increase of the carbonation rate. Unlikely, the absorbance for the case of the symmetric and antisymmetric stretching (O–H) group increases with the increase of the carbonation rate and reaches an absolute maximum value at \(X = 0.6\%\). Again, this result implies that the critical range for the variation of absorbance for the different samples occurs at \(X = 0.6\%\) for both the silicate group and the symmetric and antisymmetric (O–H) group, in good agreement with that obtained for the variation in the (RI).

In fact, increasing the rate of kinetic carbonation led to reorientation of the principal modes, which are covered by the energy state of the OPC matrix system, which in turn led to a change in the OPC matrix system. The kinetic carbonation is affected by the loss of bound water, which would lead to order–disorder (degree of order) arrangement in the OPC matrix such as the degree of polymorphism of silicate group [23,28].

**Variation of relaxation time (\(\tau\)) and the rotational energy barrier (\(U\))**

The variations of relaxation time (\(\tau\)) and the rotational energy barrier (\(U\)) with the rate of carbonation for (a) the out-of-plane (Si–O) silicate group at ≈ 520 cm\(^{-1}\), (b) the asymmetric stretching of \(\text{CO}_3^–\) mode at ≈ 1458 cm\(^{-1}\), and (c) the symmetric and antisymmetric stretching (O–H) mode at ≈ 3435 cm\(^{-1}\), are illustrated in Figure 5a, b, and c, respectively. The relaxation time (\(\tau\)) and the rotational energy barrier (\(U\)) are obtained by the help of the following equations for the relaxation time and rotational energy barrier [29–33]:

\[
\tau = 1/\pi \Delta \nu c
\]

where \(\Delta \nu\) is the full width half maximum (FWHM) of IR absorption band and \(c\) is the velocity of light and

\[
\tau = \tau_0 \exp(U/kT)
\]

where \((U)\) is the rotational energy barrier, \(k\) is the Boltzmann’s constant, \(T\) the absolute temperature, and \(\tau_0 = 10^{-11}\) s [29,30]. The relaxation time (\(\tau\)) and the rotational energy barrier (\(U\)) of the mode depend upon the chemical state of the vibrational group and the degree of freedom of motion due to it is position inside the sample during the kinetic carbonation process [34]. Figure 4a, b shows that the critical variation for the two parameters in these
variations modes is observed at $\chi = 0.6\%$, the result of which is supported by all the obtained results. The relaxation time ($\tau$) and the rotational energy barrier ($U$) of the vibrational mode depend upon the chemical state of the vibrational group, the degrees of freedom of motion, and the rate of the kinetic carbonation process in the sample. The results obtained for the relaxation time and the rotational energy barrier are in good agreement with those obtained for the RI and absorbance.

Figure 4. The variation of the absorbance with the carbonation rate for (a) the out-of-plane silicates group (Si–O) and (b) the symmetric and antisymmetric stretching of (O–H) group.
The XRD quantitative analysis for the microstructure study due to calcination and carbonation processes

The XRD of the material has always been a powerful tool for the identification of microstructure of solid materials. The XRD spectra of both the sugar-free samples CS0 and CS1 are shown in Figure 6, where the effect of the calcination temperature on the XRD peaks for the different cement hydrated phases CSH, CH, and the anhydrate clinker phases C₃S and C₂S is illustrated.
It can be shown that the peak intensity of the hydrated phase (CSH) increase while the peak intensity of the (CH) hydrated phase decrease. It can be seen that, from the calcined sugar-free sample CS1, the anhydrate phase $C_S^3S$ and $\beta-C_2S$ increases after the calcination process. The increase in the anhydrate phase after calcination can be attributed to the reaction of $SiO_2$ in silicate group ($Si-O$) with other oxides ($CaO$, $Al_2O_3$, $Fe_2O_3$, and $SO_3$) in the OPC matrix form extra amount from the clinker (dehydrated) phases [35–37]. This result is supported by the FTIR analysis of the silicate group ($\approx 520$ cm$^{-1}$) after calcination.

The general changes in the hydrated phases (i.e. CSH and CH) and the clinker dehydrated phases for OPC matrix depend on the changes of the pores system due to the evaporation of the capillary water (free water) through the dehydration of the hydrated phases and the clinker phases. The increase in the peak intensity of the (CSH) phase indicates the partial decomposition of this hydrated phase. The decrease in the intensity of CH phase is related to the recrystallization of the amorphous CH phase. This is accompanied by an increase in the intensity of the OPC matrix system [22,35,36,38].

The XRD patterns for the calcined samples (CS1–CS6) with different sugar concentrations ($0 \leq X \leq 1.1\%$) are shown in Figure 7. In this figure, it is shown that in the range ($0 \leq X \leq 0.4\%$) as the rate of carbonation (i.e. sugar

![Figure 6](image_url). The variation effect of calcination temperature (600°C). (a) Before calcinations. (b) After calcinations. (1→ CH; 2 → CSH; 3 → C3S + $\beta$C2S).
concentration) increases, the CH hydrated phase decreases due to the formation of calcite (CaCO₃). The carbonation of CSH phase depends, however, mainly on the chemical bound water and the adsorbed water on the surface of the material, where the rate of the kinetic carbonation decreases with the increase of bound water and the adsorbed water on the OPC matrix, leading to a rehydration of the hydrated phase CSH and CH. So, the carbonation does not affect the CSH hydrated phase, but it transforms it to another polymorphism in crystal structure (semi-crystalline). In the range (0.4% < X ≤ 1.1%), the rehydration process increases due to the increase in the chemical bound water in OPC system. This result is supported by the maximum height of the peak of CH phase, for X = 0.6%, in the FTIR spectra shown in Figure 2.

The calcite CaCO₃ produced during the carbonation mechanism is deposited on the pore network in hydrated cement matrix, leading to a decrease in the pore system and refinement of the porosity of the OPC

![Figure 7](image_url). The effect of kinetic carbonation for different samples (1 → CH; 2 → CSH; 3 → C3S + βC2S).
matrix (i.e. improved durability), as well as the amount of bound water. This result is supported by the variation of the \( \text{CO}_3^- \) (1420–1590 cm\(^{-1}\)) group in the FTIR analysis shown in Figure 2, where the peak has a minimum value (minimum carbonation) at \( X = 0.6\% \). Accordingly, the carbonation process of the clinker phases (\( \text{C}_3\text{S} \), and \( \beta\text{-C}_2\text{S} \)) is less than that of the hydrated phases.

For rate of carbonation (\( X > 0.6\% \)), all the XRD peaks in the diffraction pattern are decreasing due to the increase in the amorphous structure of the silica gel [23–25]. This is supported by the obtained result for the \( \text{CO}_3^- \) (1420–1590 cm\(^{-1}\)) group in the FTIR analysis besides the reduction in the bound water.

**Conclusions**

Based upon the above experimental results, the conclusions can be summarized as follows:

1. The hydrated and dehydrated phases of the OPC matrix are accompanied by clear changes due to the calcination process. The changes associated with the hydrated phases were found to be dependent on the pore system and the loss of bound water, while those associated with the dehydrated phases were found to be dependent on the activation of the silicate group to react with the other oxides in the system in order to form the \( \text{C}_3\text{S} \) and \( \text{C}_2\text{S} \) phases.

2. The carbonation process in the OPC matrix leads to dehydration of the hydrated phases (CSH and CH) through the formation of calcium carbonate (calcite) and the change in the polymorphism of CSH hydrated phase.

3. Due to the deposition of the \( \text{CaCO}_3 \) grains and occupation of the pores of the matrix, in addition to the increase of the recrystallization of the CH hydrated phase during the carbonation process, it becomes unlikely for the carbon to interact with the clinker phases.

4. Based on both the FTIR and XRD analyses, it can be concluded that the critical condition for the carbonation process in OPC matrix occurs at \( X = 0.6\% \).

5. For \( X > 0.6\% \), the carbonation process increases due to the increase in RI of the carbonyl group (C=O) and the decrease in symmetric–antisymmetric modes of (H–O) group in the context of the FTIR analysis and the decrease in the CH peaks in the context of XRD analysis. The increase in the carbonation process is attributed to the increase of the capillary porosity of the OPC matrix.

6. The increase of the carbonation process in the OPC matrix leads to more changes in the polymorphism of CSH hydrated phase and
increasing the formation of silica gel. For higher rates of carbonation, the OPC system becomes amorphous.

**Disclosure statement**

No potential conflict of interest was reported by the author(s).

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