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Original Research Article

Optimizing the Thermal Activation of Kaolinite for Sustainable Zeolite Synthesis: A Quantitative Infrared Spectroscopic Study

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Abstract

Thermal activation of kaolinite plays a critical role in determining its suitability for industrial and environmental applications, particularly for the production of reactive metakaolin. In this study, a quantitative and mechanistically informed evaluation of the amorphization and dehydroxylation behaviour of Ikere-Ekiti kaolin was conducted following calcination at temperatures between 550 and 850 °C for residence times of 15- and 120-min. Fourier-transform infrared (FTIR) spectroscopy was employed to monitor structural evolution, with emphasis on hydroxyl group removal and collapse of the kaolinite lattice. The persistence of O–H stretching bands at 3697 and 3622 cm^{-1} after calcination at 550 °C for 15 min indicates incomplete metakaolinization, whereas their disappearance at higher temperatures and longer durations confirms extensive dehydroxylation. Quantitative analysis based on spectral deconvolution and normalization reveals that amorphization is strongly dependent on both temperature and time, reaching a maximum near 650 °C after 120 min. Calcination above 750 °C results in a slight decrease in structural disorder, suggesting the onset of reorganization toward spinel-type intermediates. A schematic transformation pathway and a pseudo-Arrhenius interpretation of dehydroxylation kinetics are proposed. These results identify an optimal activation window for kaolinite and provide general insight into thermally driven transformations of layered aluminosilicates.

Keywords: kaolinite; metakaolin; amorphization; dehydroxylation; FTIR spectroscopy.

1. INTRODUCTION

Kaolin is a naturally occurring aluminosilicate clay mineral composed predominantly of kaolinite ($Al_2Si_2O_5(OH)_4$) and is widely utilized in ceramics, paper coating, pigments, construction materials, and catalysis. Upon thermal treatment, kaolinite undergoes two principal transformations: (i) dehydroxylation, which involves the removal of structural hydroxyl groups, and (ii) amorphization, characterized by the loss of long-range crystalline order and formation of metakaolin (Bish *et al.*, 2020; Gualtieri *et al.*, 2020; Madejová and Komadel, 2001). These processes significantly enhance surface reactivity and underpin the use of metakaolin as a precursor for pozzolanic binders, zeolites, and solid acid catalysts (White *et al.*, 2012).

The degree of dehydroxylation and amorphization largely determines the physicochemical reactivity of thermally activated kaolin. Insufficient thermal treatment limits reactivity, whereas excessive calcination can promote the formation of spinel or mullite phases, which reduce disorder and chemical activity (Zhou *et al.*, 2022; Sathy and Pramada, 2004; Ríos *et al.*, 2009; Yang, 2018). Therefore, identifying optimal calcination conditions remains essential for tailoring kaolin-derived materials for advanced applications.

Ikere-Ekiti (Ekiti State, Nigeria) hosts kaolin deposits with relatively high Al_2O_3 and SiO_2 contents, making them promising feedstocks for aluminosilicate synthesis. However, the kinetics and mechanistic aspects of their thermal transformation have not been comprehensively quantified. FTIR spectroscopy provides a sensitive method for tracking kaolinite transformation through changes in O–H and Si–O vibrational modes and has increasingly been applied for quantitative evaluation of structural disorder and dehydroxylation (Rahier *et al.*, 2021). In this work, FTIR spectroscopy combined with band deconvolution is employed to quantify amorphization and dehydroxylation in Ikere-Ekiti kaolin and to develop a

mechanistic framework linking aluminium coordination, reaction kinetics, and thermal energetics.

2. MATERIALS AND METHOD

2.1 Thermal activation

Raw Ikere-Ekiti kaolin was calcined in a calibrated fixed-bed furnace at 550, 650, 750, and 850 °C for residence times of 15- and 120-min. Samples were heated at a rate of 10 °C min^{-1}, maintained at the target temperature, and subsequently cooled naturally in a desiccator to minimize post-treatment rehydration.

2.2 FTIR spectroscopy

FTIR spectra were acquired using an Agilent Cary 630 spectrometer over the range 400–4000 cm^{-1} at a resolution of 1 cm^{-1}, with 32 co-added scans per sample. Pellets were prepared by thoroughly mixing 1 mg of finely ground sample with 99 mg of KBr and pressing under 12 MPa. Background spectra were recorded prior to each measurement.

2.3 Quantitative analysis

All spectra were baseline-corrected and deconvoluted using Gaussian functions in OriginPro (2023). Band areas and full widths at half maximum (FWHM) were extracted and used to calculate the amorphization index (AI), degree of amorphization (DA), and degree of dehydroxylation (DHH) following established procedures (Rahier *et al.*, 2021). In these calculations, A_t and A_0 represent the OH-band areas of treated and untreated samples, respectively.

3. RESULTS AND DISCUSSION

3.1 FTIR evolution during calcination

Raw kaolin exhibits distinct O–H stretching bands at 3697 cm^{-1} (inner-surface hydroxyls) and 3622 cm^{-1} (inner hydroxyls), confirming a well-ordered kaolinite structure (Bish *et al.*, 2020). Bands within the 1000–1100 cm^{-1} region correspond to Si–O stretching vibrations typical of layered aluminosilicates. Following calcination at 550 °C for 15 min, both hydroxyl bands remain evident, indicating incomplete dehydroxylation. Progressive attenuation and eventual disappearance of these bands at higher temperatures and longer residence times (Figure 1) confirm the formation of metakaolin, consistent with previous FTIR and in-situ studies (Cao *et al.*, 2021).

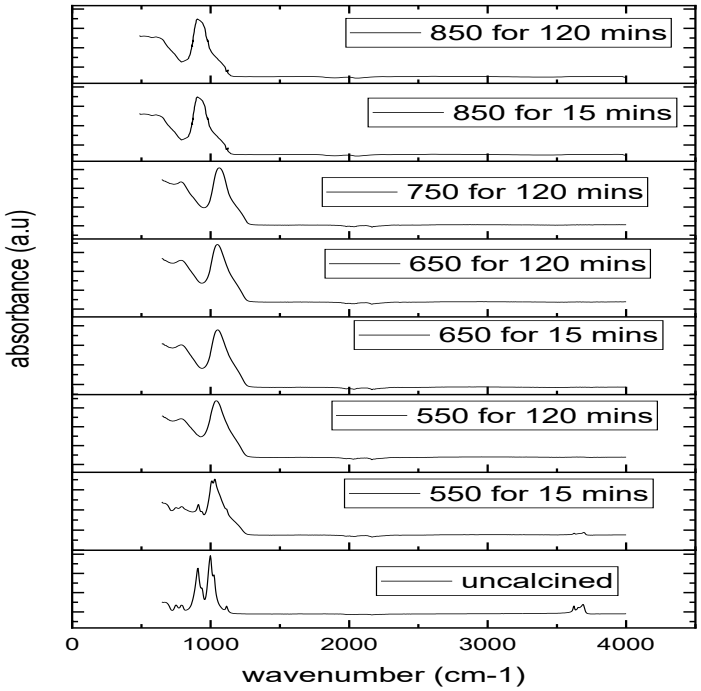

Fig. 1: FTIR spectra of heat-treated kaolin at varied temperatures and times

3.2 Quantitative amorphization behaviour

Amorphization indices derived from the FWHM of Si–O stretching bands increase with temperature up to 650 °C, indicating enhanced structural disorder. At higher temperatures, a modest decrease in amorphization is observed, suggesting partial structural reorganization. Similar trends have been attributed to the onset of short-range ordering within the amorphous aluminosilicate matrix (He *et al.*, 2022; Li *et al.*, 2023).

Amorphization Index (AI):

$$AI = \frac{FWHM_{sample}}{FWHM_{uncalcined}} \qquad 2$$

Degree of Amorphization (DA):

$$DA(\%) = \frac{AI_{sample} - AI_{uncalcined}}{AI_{max.} - AI_{uncalcined}} \quad 2.1$$

Degree of Dehydroxylation (DHH):

$$DHH(\%) = \left(1 - \frac{A_t}{A_o}\right) x100 \quad 2.2$$

where (A_t) and (A_0) are treated and untreated OH-peak areas, respectively (Rahier *et al.,* 2010)

Table 1: Amorphization parameters of Ikere-Ekiti kaolin at varied temperatures and times

Temperature (°C)	FWHM (15 min)	FWHM (120 min)	AI (15 min)	AI (120 min)	DA (%) (15 min)	DA (%) (120 min)
550	104.8	119.1	1.69	1.92	69.0	92.1
650	112.4	111.0	1.79	1.79	78.9	79.0
750	106.6	103.4	1.72	1.67	71.9	66.8
850	98.6	91.4	1.59	1.51	59.0	51.1

3.3 Degree of dehydroxylation

The degree of dehydroxylation exceeds 99 % at temperatures ≥ 650 °C, particularly for prolonged residence times. Extended calcination at 550 °C also results in near-complete dehydroxylation, highlighting the strong coupling between temperature and time. This time–temperature compensation behaviour reflects kinetically controlled hydroxyl diffusion and recombination processes (Cao *et al.*, 2021).

Table 2: Degree of dehydroxylation (DHH) of Ikere kaolin at different calcination conditions

Temperature (°C)	DHH (%) (15 min)	DHH (%) (120 min)
550	91.5	99.4
650	99.2	99.1
750	98.7	99.4
850	99.0	100

3.4 Mechanistic implications: aluminium coordination and energetics

Dehydroxylation proceeds via condensation of adjacent hydroxyl groups to form molecular water and Al–O–Al linkages, accompanied by a progressive transformation of aluminium coordination from octahedral Al(VI) to tetrahedral Al(IV) environments (Zhang *et al.*, 2024). The pronounced increase in structural disorder between 550 and 650 °C defines an effective activation window in which the apparent activation energy for hydroxyl removal is overcome. Reported activation energies for kaolinite dehydroxylation (approximately 140–200 kJ mol^{-1}) are consistent with the observed transformation behaviour (Gualtieri *et al.*, 2020; Cao *et al.*, 2021).

At temperatures ≥ 750 °C, the slight reduction in amorphization indicates initial reorganization toward spinel-type or γ-alumina-like intermediates, which precede mullite formation (Zhou *et al.*, 2022).

Al(IV) coordination The thermal evolution of Ikere-Ekiti kaolin therefore follows a three-stage pathway: (i) partial dehydroxylation of kaolinite at ≤ 550 °C; (ii) lattice collapse and formation of amorphous, highly reactive metakaolin dominated by between 550 and 650 °C; and (iii) short-range structural reorganization into spinel-type intermediates at ≥ 750 °C. This sequence may be summarized as:
kaolinite → metakaolin → spinel-type intermediate → mullite

3.5 Kinetic analysis

The kinetic barriers governing dehydroxylation can be interpreted using a pseudo-Arrhenius approach. If the degree of dehydroxylation (DHH) is treated as a proxy for reaction progress, the relationship between ln(DHH) and reciprocal temperature (1/T) reveals distinct kinetic regimes. For the 15-min calcination series, DHH shows strong temperature dependence between 550 and 650 °C, increasing from 91.5 % to 99.2 %, indicating a high apparent activation barrier. In contrast, the 120-min series exhibit reduced temperature sensitivity, with DHH values exceeding 99 % even at 550 °C. This behaviour reflects kinetic compensation; whereby prolonged thermal exposure allows the reaction to proceed despite high activation barriers. Although DHH is not a true rate constant, this analysis reinforces identification of 550–650 °C as the critical activation window for Ikere-Ekiti kaolin.

4. CONCLUSIONS

FTIR spectroscopy provides a robust quantitative framework for evaluating amorphization and dehydroxylation in Ikere-Ekiti kaolin. Maximum structural disorder and near-complete hydroxyl removal are achieved within a narrow activation window of 550–650 °C, while prolonged residence time enables complete transformation at lower temperatures. Calcination above 750 °C promotes partial recrystallization, reducing amorphous character. These findings offer mechanistic and kinetic insight applicable

to kaolin resources beyond the studied deposit and support optimized thermal activation strategies for metakaolin-based materials.

While FTIR spectroscopy enables sensitive monitoring of structural disorder, complementary techniques such as solid-state NMR and in-situ thermal analysis would permit direct quantification of aluminium coordination populations and activation energies. Future studies integrating multi-technique approaches across kaolins of varying crystallinity will further refine universal activation guidelines.

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