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BENEFICIATION AND PROCESSING OF NATURAL AND TECHNOGENIC RAW MATERIALS

Research paper

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**Characterization and thermal behavior of some types of kaolin of different origin from Northern Vietnam**

T. T. T. Nguyen , H. B. Bui

Hanoi University of Mining and Geology, Hanoi, Vietnam nguyenthithanhthao@humg.edu.vn**Abstract**

Kaolin (mainly composed of kaolinite, whose chemical formula is $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$), serves as a versatile raw material widely used in various industries including production of ceramics, paper, paints, cosmetics, pneumatics, building materials, and hazardous waste storage. In the northern part of Vietnam, due to favorable geological conditions, there are diverse deposits of high quality kaolin of different origin and scale. Decades of research indicate the diversity of kaolin sources in the region, with special attention paid to hydrothermally altered and exchange types of kaolin, the formation of which is associated with complex processes of weathering, hydrothermal alteration and reprecipitation. The aim of this study was to characterize three different types of kaolin derived from different sources in Northern Vietnam (from weathered pegmatites, weathered felsic effusives, and hydrothermal-metasomatic altered rocks). The main focus was to analyze the thermal behavior of these samples during calcination in the temperature range from 300 °C to 1,100 °C. The comprehensive characterization was performed by X-ray diffraction (XRD), FT-IR spectroscopy (FT-IR), thermal analysis (thermogravimetry/differential thermogravimetry (TG/DTG)) and scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDS). The results showed that kaolinite with particle size less than 2 μm was identified in all samples. Minor amounts of muscovite and montmorillonite are present in some samples, and pyrophyllite is present in a sample from the hydrothermally altered rocks. Kaolinite morphology in all the samples showed typical forms including hexagonal and pseudohexagonal. The main chemical constituents of the samples are SiO_2 and Al_2O_3 ; in addition to these, $\text{K}_2\text{O} + \text{Na}_2\text{O}$, TiO_2 and iron are present in smaller quantities. Thermal analysis allowed to reveal the formation of metakaolinite phase at temperatures around 494 °C and 507 °C in the two studied samples from weathered rocks, while the pyrophyllite-bearing sample undergoes this transition at a higher temperature of 653.8 °C. The onset of metakaolinization was observed at about 500 °C for the weathered rock samples and about 700 °C for the pyrophyllite-bearing sample. In addition, mullitization leading to the formation of mullite was evident at 1,100 °C. The study findings allow concluding that the studied kaolins can be used in traditional ceramics production.

Keywords

kaolin, $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$, pyrophyllite, mullite, thermal analysis, metakaolinite, mullitization, pegmatite, Northern Vietnam

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ОБОГАЩЕНИЕ, ПЕРЕРАБОТКА МИНЕРАЛЬНОГО И ТЕХНОГЕННОГО СЫРЬЯ

Научная статья

Характеристика и термическое поведение некоторых видов каолина различного происхождения из Северного Вьетнама

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Ханойский университет горного дела и геологии, г. Ханой, Вьетнам nguyenthithanhthao@humg.edu.vn**Аннотация**

Каолин (состоящий в основном из каолинита, химическая формула которого $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) служит универсальным сырьем, широко используется в различных отраслях промышленности, включая производство керамики, бумаги, красок, косметики, пневматики, строительных материалов и хранение



опасных отходов. В северной части Вьетнама благодаря благоприятным геологическим условиям находятся разнообразные месторождения высококачественного каолина различного происхождения и масштаба. Хотя в ряде работ изучены качество, потенциал, распространение и происхождение типов каолина в Северном Вьетнаме, исследования различий между каолинами из разных источников весьма ограничены. Целью данного исследования было определение характеристик трех различных типов каолина, полученных из различных источников в Северном Вьетнаме (из выветренных пегматитов, выветренных изверженных магматических пород кислого состава и гидротермально-метасоматических измененных пород). Основное внимание было уделено анализу термического поведения этих проб в ходе прокалывания в диапазоне температур от 300 до 1100 °С. Всесторонняя характеристика проводилась методами рентгеноструктурного анализа (XRD), Фурье-ИК-спектроскопии (FT-IR), термического анализа (термогравиметрия/дифференциальная термогравиметрия (TG/DTG)) и сканирующей электронной микроскопии с энергодисперсионной рентгеновской спектроскопией (SEM-EDS). Результаты показали, что во всех пробах был обнаружен каолинит с размером частиц менее 2 мкм. В отдельных пробах присутствуют незначительные количества мусковита и монтмориллонита, а в пробе из гидротермально измененных пород – пиррофиллита. Морфология каолинита во всех пробах проявлялась в типичных формах, включая гексагональную и псевдогексагональную. Основными химическими компонентами являются SiO_2 и Al_2O_3 ; помимо них, в меньших количествах присутствуют $\text{K}_2\text{O} + \text{Na}_2\text{O}$, TiO_2 и общее железо. Термический анализ выявил образование метакаолинитовой фазы при температурах около 494 и 507 °С в двух изученных пробах из выветренных пород, а пиррофиллитсодержащая проба претерпевает этот переход при более высокой температуре – 653,8 °С. Начало метакаолинитизации наблюдалось при температуре около 500 °С для проб из выветренных пород и около 700 °С – для пиррофиллитсодержащей пробы. Кроме того, при 1100 °С проявилась муллитизация, приводящая к образованию муллита. Полученные результаты позволяют сделать вывод о возможности применения этих проб каолина в традиционном производстве керамики.

Ключевые слова

каолин, $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$, пиррофиллит, муллит, термический анализ, метакаолинит, муллитизация, пегматит, Северный Вьетнам

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Introduction

Kaolin (mainly composed of kaolinite, whose chemical formula is $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$), serves as a versatile raw material widely used in various industries including production of ceramics, paper, paints, cosmetics, pneumatics, building materials, and hazardous waste storage [1–3]. The quality and applicability of kaolin depends on factors such as its chemical composition, physical properties, mineralogical composition and structural morphology. For example, Maja (2017) showed that clay from Slatina deposit in Serbia has composition and other characteristics suitable for ceramic and construction industries, especially for the production of tiles, thin-walled hollow bricks, and roof tiles/lightweight blocks [4]. Further studies aimed at investigating properties such as purity, mineralogical composition, color and texture by researchers such as Hernández et al. (2019) identified the characteristics of kaolin deposits in Venezuela, suggesting potential demand for this kaolin to be processed for using in the pharmaceutical industry [5]. A number of researchers emphasize the need for a comprehensive study of the physicochemical properties of kaolin raw materials and their behavior under calcination conditions before the use in practice.

In the northern part of Vietnam, due to favorable geological conditions, there are diverse deposits of high quality kaolin of different origin and scale. Decades of research indicate the diversity of kaolin sources in the region, with special attention paid to hydrothermally altered and exchange types of kaolin, the formation of which is associated with complex processes of weathering, hydrothermal alteration and reprecipitation [6, 7]. Although several works have studied the quality, potential, distribution, and origin of kaolin types in North Vietnam [8, 9], studies on the differences between kaolins from different sources are very limited.

This paper presents the application of combined analytical techniques such as X-ray diffraction analysis (XRD), Fourier transform infrared spectroscopy (FT-IR), thermal analysis (thermogravimetry/differential thermogravimetry (TG/DTG)), and scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDS) to investigate the characteristics of kaolin raw materials of different origins from several mines in Northern Vietnam, as well as their properties/behavior under calcination conditions. The results obtained allowed to comprehensively and more completely assess the qualitative characteristics of the kaolins and contribute to their more efficient application.

Review of geological characteristics of some kaolin types at mines in Vietnam

Kaolin from weathered pegmatites

Kaolin formed in weathered pegmatites is widely distributed in the Lao Cai area in Northern Vietnam [10]. These kaolin-bearing bodies are usually cylinder-shaped and have a branched morphology. Pegmatite bodies of various sizes are distributed in metamorphic formations of Proterozoic and Lower Paleozoic age. The thickness of the kaolin body depends on the terrain. In a vertical cross-section, the pegmatite bodies are stratified: the uppermost layer consists of kaolin, followed by a layer of low-weathered pegmatites, and the lowest layer is represented by fresh pegmatites. This type kaolin mines are usually medium to small scale mining operations. This variety of kaolin is usually fine-grained, rich in aluminum and characterized by relatively high iron content, often has a yellow or dark yellow hue. The -0.21 mm undersize kaolin recovery ranges from 30 to 60%, averaging below 40%. Fig. 1 shows a geological cross-section of a kaolin body of such nature.

Kaolin from weathered felsic effusive rocks

Kaolin formed in weathering crust in effusive rocks is widely distributed throughout Northern Vietnam in different structural zones [11]. This type of kaolin deposits, usually formed in the weathering crust in rhyolite and rhyolite-porphyry rocks, is characterized by small-scale occurrences, often hopper-shaped and lens-shaped. Kaolin of this type is usually fine-grained, present in white or pinkish-white tints. The -0.21 mm undersize kaolin recovery ranges from 50 to 90%, averaging about 70%. Fig. 2 shows a cross-section of a deposit of this type of kaolin.

Kaolin of hydrothermal-metasomatic genesis

Kaolin bodies containing pyrophyllite (hereinafter referred as kaolin-pyrophyllite bodies) are the product of contact alteration processes involving hydrothermal solutions and various rocks such as rhyolites, rhyolite porphyries, felsites and tuffs [12]. These bodies are intersected by faults or, conversely, are accommodated by fault zones, identified at a mine, that determines their relatively large scale.

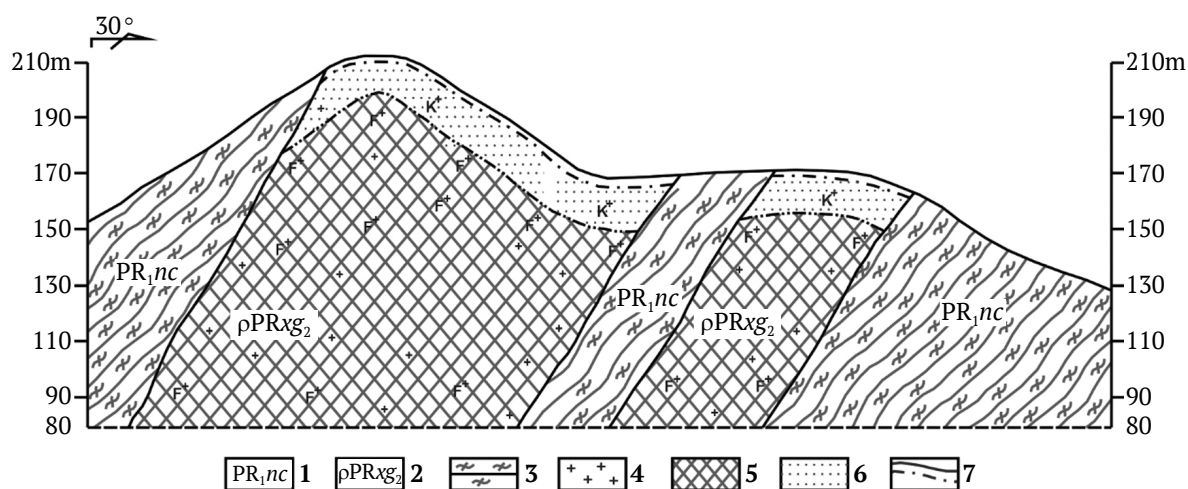


Fig. 1. Geological cross-section at Son Man kaolin mine, Lao Cai Province:

1 – quartz schists, mica, gneisses, interbedded quartzites; 2 – aplite and pegmatite veins; 3 – mica crystalline schists; 4 – fresh (unweathered) pegmatites; 5 – low-weathered pegmatites; 6 – kaolin; 7 – topsoil (land cover)

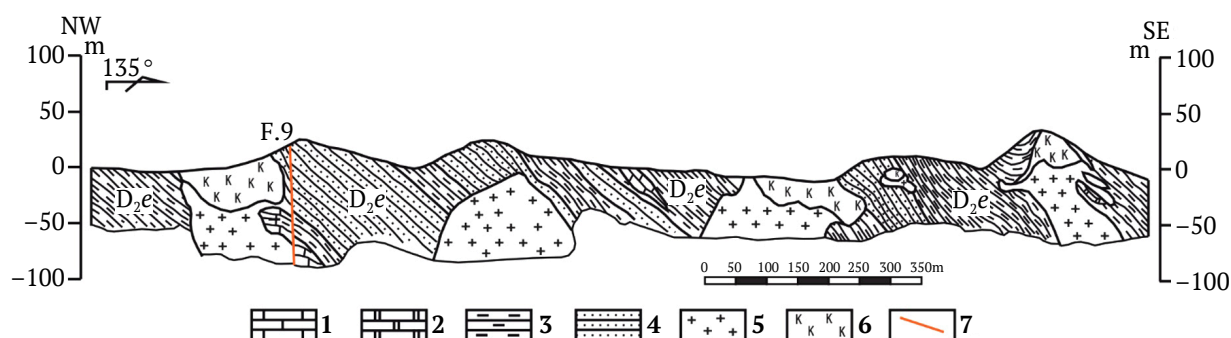


Fig. 2. Geological cross-section at Minh Tan kaolin mine, Hai Duong Province:

1 – quaternary sediments; 2 – limestone; 3 – silicified limestone; 4 – shale, sandstone; 5 – keratophyre; 6 – kaolin; 7 – fault

The morphology of such kaolin-pyrophyllite bodies is complex, with irregular randomly manifested bulges. Fig. 3 shows a cross-section of a kaolin-pyrophyllite body for better visualization.

Research Methodology

Sample Preparation

Bulk samples were collected from the mining zones of three kaolin mines, kaolins of which have different genesis (type): kaolin from the weathering zone of pegmatites of the Thach Khoan complex at Son Man mine, Lao Cai Province (kaolin from weathered pegmatites); kaolin from the weathering zone of rhyolites of the Binh Lieu Formation, Minh Tan mine, Hai Duong Province (kaolin from weathered effusives), and kaolin-pyrophyllite of hydrothermal-metasomatic genesis from Tan Mai mine, Quang Ninh Province (kaolin-pyrophyllite) (Figs. 1–3). These natural kaolin samples were pulverized, dissolved, mixed, and screened using a sieve with mesh size $< 63 \mu\text{m}$. The screening undersizes were placed into laboratory bags and used for subsequent analyses and tests. For the identification of clay minerals, a clay fraction with particle size $< 2 \mu\text{m}$ was obtained by decantation. The particle size fraction $< 2 \mu\text{m}$ was used to prepare cylindrical pellets for further studies. They were further subjected to various treatments including air drying, glycolation with ethylene glycol, and heating to 350°C .

The cylindrical pellets were made by uniaxial dry pressing of the samples at a pressure of 40 MPa. The cylindrical pellets were then dried at 60°C for 24 h in an oven. The dried cylindrical pellets were heated to selected temperatures of 300, 500, 700, 900 and 1100°C at a heating rate of $5^\circ\text{C}/\text{min}$ using an electric laboratory furnace. For each selected temperature,

the sample was calcined for 2 h and “quenched” to room temperature under ambient conditions to avoid crystallization of amorphous metakaolinite. The portions of the heated samples were ground using an agate mortar and pestle for subsequent analyses.

Sample characterization

The morphological properties of minerals and the mineralogical composition of the samples were investigated using a scanning electron microscope (SEM – Quanta 450) with energy dispersive X-ray spectroscopy (EDS). The mineralogical analysis of the samples was performed by X-ray diffraction (XRD) analysis. The X-ray diffraction patterns of the samples processed under different conditions were also obtained on a Siemens model D5005 powder X-ray diffractometer with Cu-K α radiation at 40 kV and 30 mA, scanning from 3 to 70° at an angular velocity ω (2θ in the Figures below) of $2^\circ/\text{min}$. FT-IR spectra were recorded between $4,000$ and 400 cm^{-1} with a resolution of 2 cm^{-1} using a Shimadzu IR Prestige-21 spectrometer. The thermal behavior of each sample was determined using the techniques of thermogravimetry/differential thermogravimetry (TG/DTG) in nitrogen atmosphere in the range from room temperature up to $1,100^\circ\text{C}$ at a heating rate of $10^\circ\text{C}/\text{min}$. The nitrogen adsorption and desorption isotherms of the kaolin samples heat-treated at 196°C were obtained using a Micromeritics ASAP 2020a instrument.

Findings and Discussion

Characterization of natural kaolin materials

X-ray diffraction analysis

Fig. 4 shows X-ray diffraction patterns of three natural kaolin materials (the samples: kaolin from weathered pegmatites, kaolin from weathered felsic

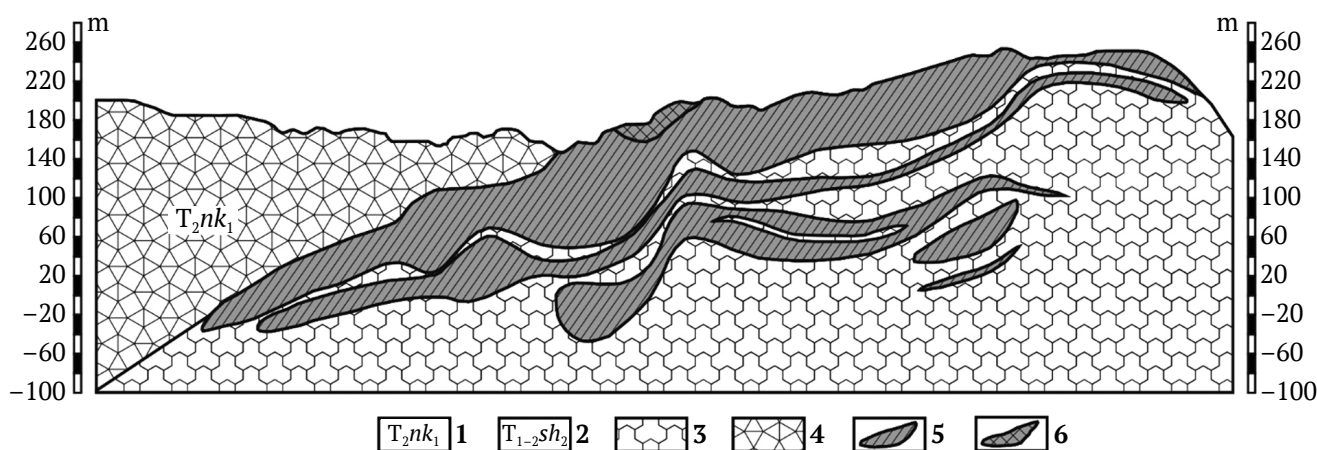


Fig. 3. Geological cross-section at a kaolin-pyrophyllite mine in Tan Mai, Quang Ninh Province:

1 – Na Khuat Formation; 2 – Khon Lang Formation; 3 – rhyolite-dacite tuff;
4 – siltstone; 5 – alunite; 6 – kaolin-pyrophyllite

effusives, and kaolin-pyrophyllite) with a particle size of 2 μm subjected to different test conditions: aged at room temperature, treated with ethylene glycol, and heated at 350 °C. These XRD patterns consistently show the predominance of kaolinite in all three samples, characterized by well-defined peaks at 7.18 Å, 4.48 Å and 3.58 Å [1]. In addition, relatively weak basal reflections at 10.0 Å, 5.02 Å and 3.35 Å indicate the presence of muscovite in the kaolin samples from pegmatites and the kaolin samples from effusives [13]. It is important to note that pyrophyllite is clearly identified in the kaolin-pyrophyllite sample obtained from Tan Mai mine, as evidenced by characteristic peaks of 9.2 Å, 4.59 Å, 4.14 Å, 3.79 Å, and 3.07 Å [14]. After the treatment with ethylene glycol and heating at 350 °C, the characteristic peaks corresponding to kaolinite, muscovite, and pyrophyllite remain on the X-ray diffraction patterns. However, in the case of the kaolin from weathered effusives sample, a noticeable shift of the peak associated with montmorillonite from 15.1 Å to 16.9 Å after ethylene glycol treatment was observed. This observation shows that, in addition to the predominance of kaolinite of < 2 μm size fraction in all the samples, the kaolin from weathered pegmatites is characterized by the presence of muscovite, the kaolin from weathered effusives is accompanied by muscovite and montmorillonite, and the kaolin-pyrophyllite sample consists predominantly of pyrophyllite. These findings emphasize that different conditions during kaolin formation can lead to a variety of mineralogical compositions, which can influence the calcination processes and their outcomes.

Analysis by scanning electron microscopy and energy dispersive X-ray spectroscopy techniques

Scanning electron microscopy (SEM) images of the minerals of the kaolin samples (the kaolin from weathered pegmatites (a), the kaolin from weathered effusives (b) and the kaolin-pyrophyllite (c)), are shown in Fig. 5. These images clearly show the characteristic morphology of kaolinite with a hexagonal thin-plate structure. The mineral grains are intricately attached together, displaying their distinctive characteristics. The results of EDS (energy dispersive X-ray spectroscopy) analysis shed light on the mineral elemental compositions. Silicon oxide (SiO_2) and aluminum oxide (Al_2O_3) were found as major components, which correspond to the expected presence of silicon (Si) and aluminum (Al) in the chemical formula of kaolinite, $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$. Table 1 shows the average chemical composition of the three kaolin samples (semi-quantitative determination by EDS analysis).

Fourier-transform infrared (FT-IR) spectroscopy analysis

Fourier-transform infrared (FT-IR) analysis showed the presence of various functional groups in the studied samples (Fig. 6). Notably, the absorption peaks at 3,687 and 3,619 cm^{-1} are associated with the stretching (bond) vibrations of O–H group bond. In addition, the bands at 1,114 and 684 cm^{-1} correspond to the stretching vibrations of Si–O bond, and the absorption bands at 1,034 and 998 cm^{-1} refer to the stretching vibration region of Si–O–Si bond. The band at 909 cm^{-1} corresponds to the stretching vibrations of Al–OH bond. These bands of stretching vibrations are

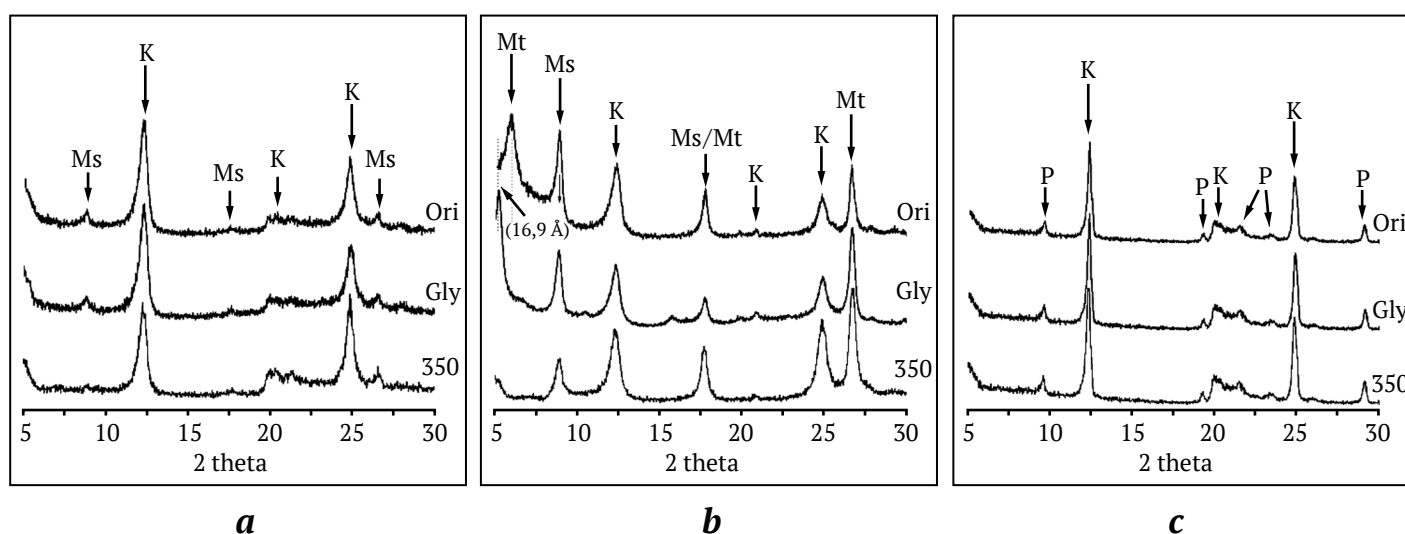


Fig. 4. X-ray diffraction pattern (XRD) of three samples (kaolin from weathered pegmatites (a), kaolin from weathered effusives (b) and kaolin-pyrophyllite (c)) with particle size < 2 μm under different test conditions: At room temperature (Ori); treatment with ethylene glycol (Gly); heating at 350 °C (350): K = kaolinite, Ms = muscovite, P = pyrophyllite and Mt = montmorillonite (in Figures hereinafter the kaolin from weathered effusives is denoted as K-Mag)

characteristic of kaolinite, indicating the significant presence of kaolinite in the samples [15–17]. Furthermore, the possible presence of quartz, in addition to kaolinite, is indicated by the bands detected in the samples at 789, 753, and 592 cm^{-1} .

Behavior of three samples under thermal action

Thermal analysis

The results of thermogravimetry/differential thermogravimetry (TG/DTG) of three samples (the kaolin from weathered pegmatites (*a*), the kaolin from weathered effusives (*b*), and the kaolin from altered rocks containing pyrophyllite (*c*)) are presented in Fig. 7. The thermal curves in Fig. 7 show the peaks of different processes (endothermic and exothermic processes) during heating. The endothermic processes occurring at low temperatures (around 79.7, 87.2, and 73.8 $^{\circ}\text{C}$) were desorption of surface H_2O and dehy-

dratation. The formation of metakaolinite phase is indicated by endothermic peaks at 494.8 $^{\circ}\text{C}$ for the kaolin from weathered pegmatite sample, 507.1 $^{\circ}\text{C}$ for the kaolin from weathered effusive sample, and 653.8 $^{\circ}\text{C}$ for the kaolin-pyrophyllite sample. The difference in the temperature of the endothermic peaks between the samples may be due to the differences in minerals morphology, composition and grain size. The kaolin-pyrophyllite sample has the highest endothermic peak temperature (653.8 $^{\circ}\text{C}$) among all the three samples, indicating the natural stability of kaolin of hydrothermal-metasomatic genesis. Analysis of the DTG data in Fig. 7 indicates a change in the weight of the tested materials during the heating process. The weight reductions were 9.77, 11.8, and 13.9% for the samples of the kaolin from weathered pegmatite, the kaolin from weathered effusives, and the kaolin-pyrophyllite, respectively.

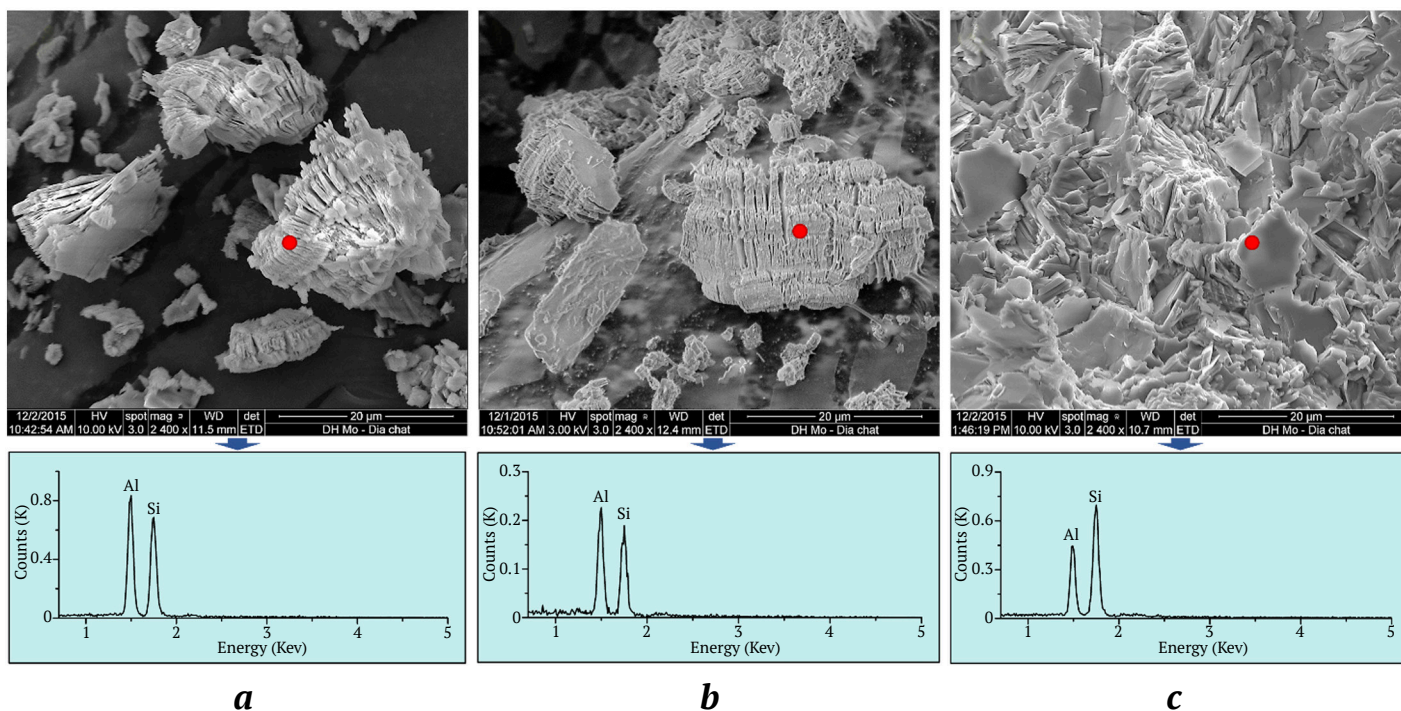


Fig. 5. SEM images and EDS results for three samples (kaolin from weathered pegmatites (*a*), kaolin from weathered effusives (*b*), and kaolin-pyrophyllite (*c*))

Chemical composition of three kaolin samples (EDS results)

Table 1

Sample	Chemical composition, % (from – to)							
	Na_2O	MgO	Al_2O_3	SiO_2	K_2O	CaO	TiO_2	Fe_2O_3
Kaolin from weathered pegmatites (<i>a</i>)	0.05–0.27	0.33–0.59	42.8–46.5	50.2–52.2	0.51–0.99	0.11–0.21	0.11–0.19	0.09–0.58
Kaolin from weathered effusives (<i>b</i>)	0.06–1.63	0.12–0.35	13.6–20.1	65.7–75.7	2.51–5.22	0.08–1.11	0.03–0.11	0.52–1.96
Kaolin—pyrophyllite (<i>c</i>)	0.21–1.30	0.05–0.56	10.5–38.6	11.5–89.3	0.16–1.20	0.05–1.42	0.05–1.35	0.03–2.51

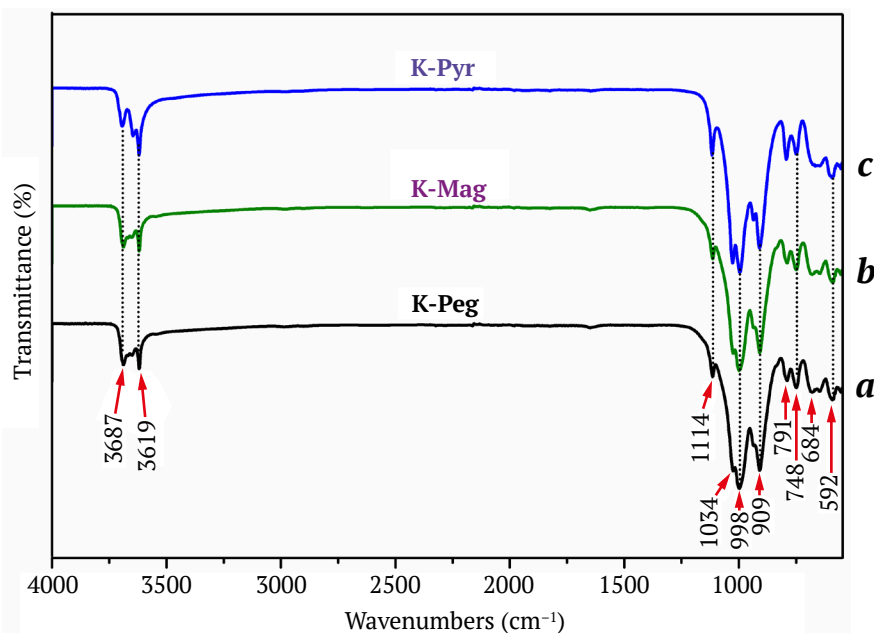
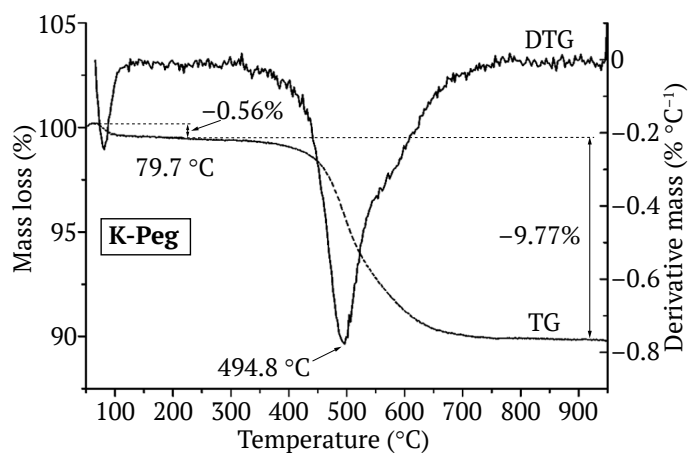
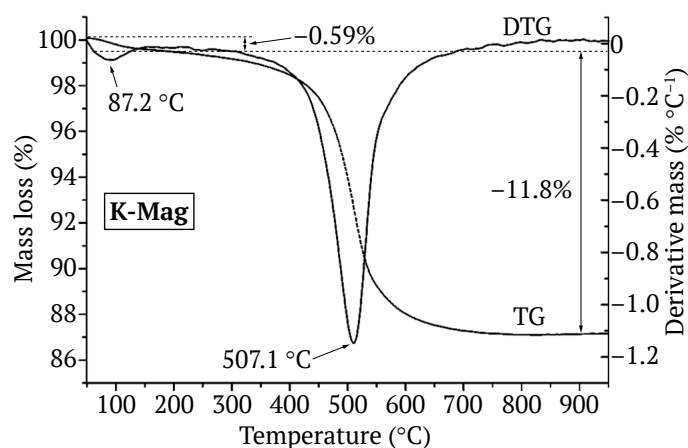


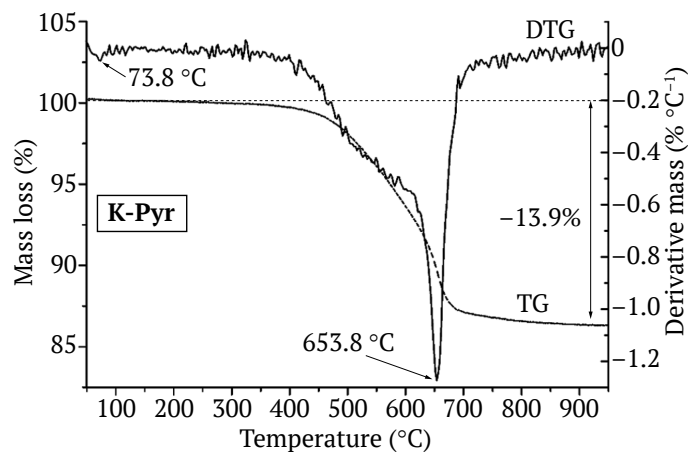
Fig. 6. FT-IR spectroscopy patterns of three samples (kaolin from weathered pegmatites (a), kaolin from weathered effusives (b) and kaolin-pyrophyllite (c))



a



b



c

Fig. 7. Analysis of thermogravimetry/differential thermogravimetry (TG/DTG) study results for three samples (the kaolin from weathered pegmatites (a), the kaolin from weathered effusives (b), and the kaolin-pyrophyllite (the kaolin from altered rocks containing pyrophyllite) (c))

X-ray diffraction analysis

Fig. 8 shows the X-ray diffraction patterns of three samples (the kaolin from weathered pegmatites, the kaolin from weathered effusives, and the kaolin-pyrophyllite) at different temperatures (300, 500, 700, 900, and 1,100 °C). The analysis shows that the kaolin from weathered pegmatite sample consists mainly of kaolinite, but also quartz and muscovite are present. In contrast, the kaolin-pyrophyllite sample consists mainly of kaolinite and pyrophyllite, with minor quartz present. Over the entire temperature range from room temperature to 500 °C, the peaks in the X-ray diffraction (XRD) patterns corresponding to these minerals remain relatively stable, although they lose in intensity while the temperature increases. At 700 °C kaolinite peaks disappear in all the samples, while those of quartz and pyrophyllite remain. The disappearance of kaolinite peaks indicates transformation into metakaolinite characterized by amorphous structure, which is indicated by wide “bumps” on the X-ray diffraction patterns. Notably, the weak XRD peak of pyrophyllite at 700 °C emphasizes its thermal stability that is consistent with the thermal analysis findings and the hydrothermal-metasomatic genesis of pyrophyllite. The most significant changes in the X-ray diffraction patterns occur at 1,100 °C, which mean melting and destruction of the structures of the initial minerals with the formation of mullite,

a new mineral phase. This conclusion is consistent with the observations of previous studies [18, 19] emphasizing the dependence of mineralogical composition transformation during calcination on the initial mineralogical composition of samples. The presence of pyrophyllite in the kaolin-pyrophyllite sample markedly influences the phase transition, leading to the formation of new minerals.

SEM analysis

SEM images of three samples: kaolin from weathered pegmatites, kaolin from weathered effusives, and kaolin-pyrophyllite subjected to calcination at temperatures of 500 °C, 700 °C, 900 °C и 1100 °C are presented in Fig. 9. At 500 °C, a distinct minerals morphology is observed and mineral grain boundaries in all three samples remain discretely defined, indicating minimal impact on structural integrity and chemical bonds at this temperature. However, when the heating temperature is increased from 700 to 1,100 °C, noticeable transformations occur. In the interval from 700 to 900 °C, the morphology of minerals undergoes a gradual change, showing signs of melting. The differences between the initial substances in the samples begin to disappear. In particular, the SEM images at 1,100 °C clearly show melting manifestations, indicating that melting of the materials hides the initial boundaries and morphology of

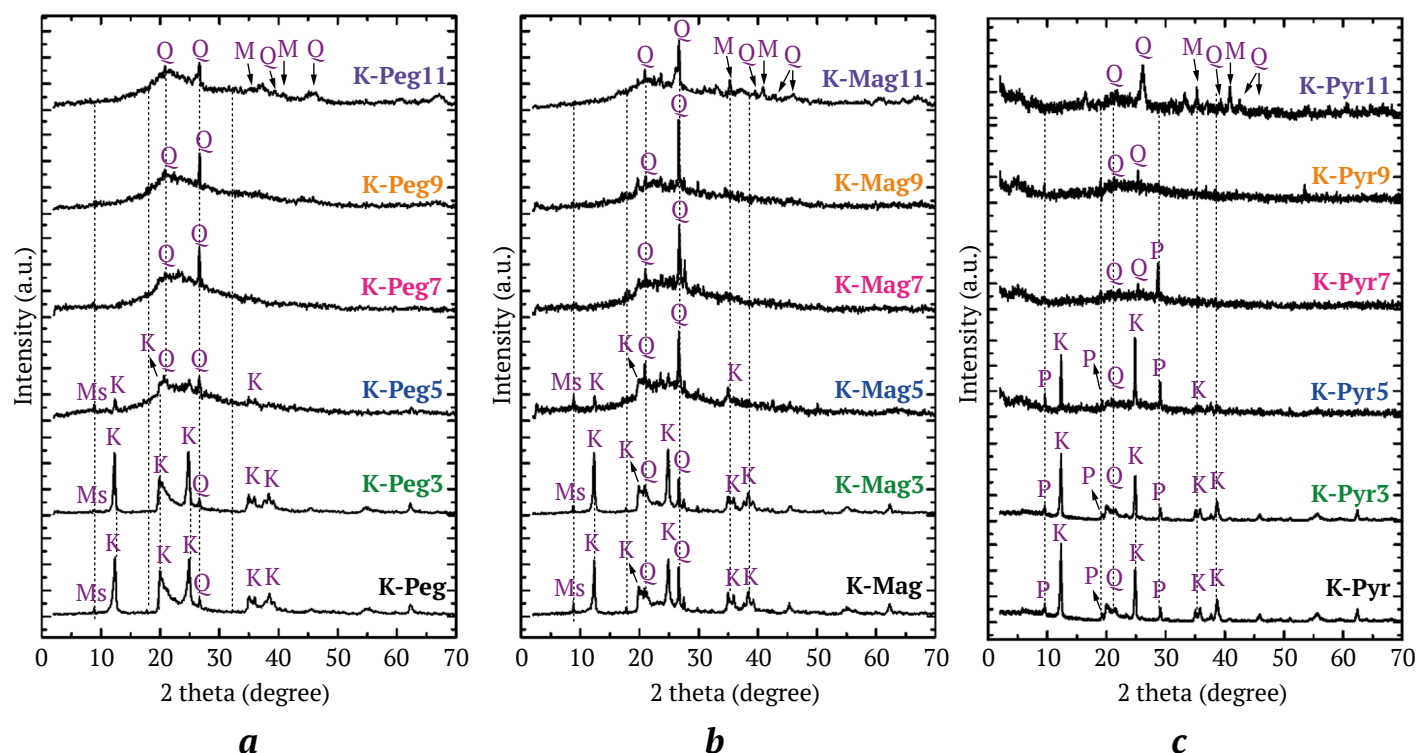


Fig. 8. X-ray diffraction patterns (XRD) of three samples (kaolin from weathered pegmatites (a), kaolin from weathered effusives (b), and kaolin-pyrophyllite (c)) at different temperatures (300 °C (3), 500 °C (5), 700 °C (7), 900 °C (9) и 1,100 °C (11)): K = kaolinite, Ms = muscovite, P = pyrophyllite, Q = quartz, M = mullite

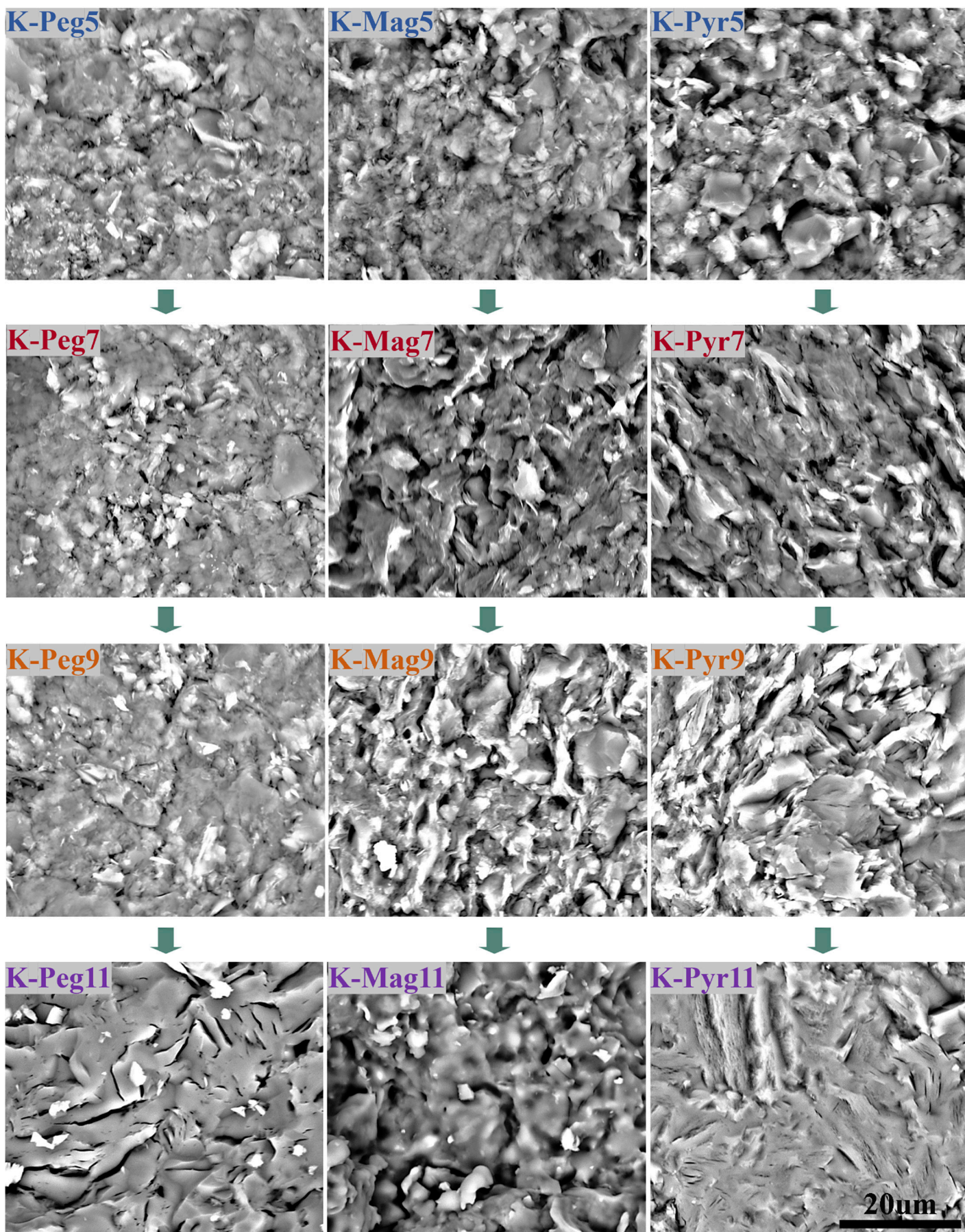


Fig. 9. SEM images of three samples (kaolin from weathered pegmatites, kaolin from weathered effusives, and kaolin-pyrophyllite) at different temperatures (500 °C (5), 700 °C (7), 900 °C (9), and 1100 °C (11))



the mineral grains. In the case of the kaolin-pyrophyllite sample, characterized by increased stability, the pyrophyllite morphology shows certain stability in the temperature range from 700 to 1,100 °C. The observed morphological changes in the samples at different calcination temperatures, as seen in the SEM images, are consistent with the other research findings, including XRD and FT-IR spectroscopy.

Conclusion

In this study, the fundamental characteristics of three types of kaolin obtained from different sources and of different genesis (from weathered pegmatites, weathered felsic effusives, and hydrothermal-metasomatic rocks), were evaluated using X-ray diffraction analysis (XRD), FT-IR spectroscopy (FT-IR), thermal analysis (thermogravimetry/differential thermogravimetry (TG/DTG)) and scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDS). The studies showed that kaolinite was the predominant mineral in all samples characterized by particle sizes less than 2 µm. In addition, the sample of kaolin from weathered pegmatite contains insignificant amounts of muscovite, and the sample of kaolin from weathered effusives contains montmorillonite. It is noteworthy that the

kaolin sample of hydrothermal-metasomatic genesis contains a relatively significant amount of pyrophyllite. The main chemical constituents of the kaolin samples are SiO₂ and Al₂O₃; in addition to these, K₂O + Na₂O, TiO₂ and iron are present in smaller quantities. Thermal analysis revealed phase transitions at different temperatures, with the metakaolinite phase formation around 494 and 507 °C in the kaolin samples from weathered pegmatites and weathered felsic effusives, respectively. The pyrophyllite-bearing kaolin sample (of hydrothermal-metasomatic origin) undergoes this phase transition at a higher temperature of 653.8 °C that indicates its natural stability. To evaluate the calcination behavior of the kaolins, the samples were pressed at a pressure of 40 MPa and calcined at temperatures ranging from 300 to 1,100 °C. XRD and SEM analyses showed that metakaolinization starts around 500 °C in the kaolin samples from weathered pegmatites and weathered effusives, and around 700 °C in the pyrophyllite-bearing kaolin sample. The process of mullitization becomes evident at 1,100 °C, leading to the formation of a new mineral, mullite. The study findings presented here have key implications for the production of traditional ceramics using natural kaolin materials of different origins.

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