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Analysis of clay characteristics based on illite/smectite interstratified minerals

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Abstract: With the booming development of the cosmetics industry, consumers have increasingly stringent requirements for the quality and efficacy of cosmetics, prompting researchers to explore new, efficient and safe raw materials. As a special member of the clay mineral family, illite/smectite interstratified minerals are formed by orderly or disorderly stacking of illite and smectite layers in a specific proportion. They are of great significance in the fields of geology for diagenetic history restoration and hydrothermal alteration research, especially closely related to the formation, migration and storage of petroleum. However, research on illite/smectite interstratified minerals in the cosmetics field is still in its infancy.This study is based on the Xuefeng mud from Xuefeng Mountain in Hunan Province, China, and conducts multi-dimensional analyses. It delves deeply into aspects such as crystal structure, chemical composition, and external properties, including the clay characteristics of illite/smectite interstratified minerals and their differences from illite. The study finds that the main component of Xuefeng mud is illite/smectite interstratified minerals with a relatively low mixing ratio. In terms of chemical composition, the proportions of elements such as Fe and K are much higher than those in illite, and the isomorphism ratio is extremely high, which is consistent with the characteristics of illite/smectite interstratified minerals. There are obvious differences from illite in particle size distribution, oil contact angle, viscosity, powder characteristics, and other aspects.In addition, the Xuefeng mud containing illite/smectite interstratified minerals does not exhibit the significant water absorption and swelling phenomenon of smectite, eliminating the greasy feeling. It can endow Xuefeng mud with a delicate, smooth, refreshing, and comfortable touch, optimize the adhesion of cosmetics, and enhance the delicacy and durability of makeup. Analyzing the clay characteristics of Xuefeng mud, whose main component is illite/smectite interstratified minerals, can help understand the particularities of illite/smectite interstratified minerals, guide their application in the cosmetics industry, and has certain reference significance for exploring natural and green mineral raw materials for cosmetics.

1. Introduction

With the booming development of the cosmetics industry, consumers' requirements for the quality and efficacy of cosmetics are increasingly stringent, prompting researchers to constantly explore efficient, and safe raw materials. Clay minerals have attracted much attention in the cosmetics field due to their unique physical and chemical properties, such as adsorption, ion exchange, and dispersibility, providing many possibilities for product innovation.[1.2]

Illite/smectite interstratified minerals, as special members of the clay mineral family, are formed by the orderly or disorderly stacking of illite and smectite lamellae in a specific proportion.[3-5] They inherit some excellent properties of illite and smectite and exhibit unique properties due to their special structure. In the field of geology, they are of great significance for reconstructing diagenetic history and studying hydrothermal alteration, especially in petroleum geology, where they are closely related to the formation, migration, and storage of petroleum. However, in the cosmetics field, the research on illite/smectite interstratified minerals is still in its infancy, and many of their properties and application potentials remain to be explored.[6,7] In-depth exploration of the characteristics of illite/smectite interstratified minerals can not only fill the gap in the research of cosmetics raw materials but also provide innovative ideas for the development of new cosmetics and expand the application boundary of clay minerals in non-geological fields, which has important theoretical and practical significance. By systematically studying their structure, composition, and physical and chemical properties, it is expected to develop cosmetics with excellent adsorption and cleaning, moisturizing and nourishing, oil control, and anti-acne effects, meeting the diverse needs of consumers and promoting the high-quality development of the cosmetics industry.[8,9]

In the field of geology, the research on illite/smectite interstratified minerals has been quite in-depth. Many scholars have focused on their formation mechanism. Since the 1930s, foreign scholars proposed that potassium-rich minerals such as feldspar and mica decompose to provide the material basis for the transformation of smectite to illite.[10-12] With the change of burial depth and temperature, smectite dehydrates, and ion substitution promotes K^+ to enter the crystal layer to form illite/smectite interstratified minerals. In terms of structure analysis, according to the stacking rules of unit crystal layers, illite/smectite interstratified minerals are divided into regular (orderly) and irregular (disorderly) mixed-layer clay minerals.[13,14] The degree of orderliness has become a key indicator of the transformation degree from smectite to illite. The higher the degree of orderliness, the deeper the transformation degree. For the calculation of the mixed-layer ratio, many methods have been developed abroad, such as Weaver's curve method and Reynolds' method.[15,16]

However, in the cosmetics field, the research on illite/smectite interstratified minerals is still in its initial stage. Currently, only a small number of studies have focused on their potential applications in cosmetics, such as using their adsorption properties to clean the skin and their ion exchange properties to regulate the skin microenvironment. However, these explorations mostly remain at the stage of theoretical assumptions and preliminary experiments, lacking systematic and in-depth research. There is no comprehensive and accurate analysis of the specificity of illite/smectite interstratified minerals and their differences from illite.[17] On this basis, this study deeply investigated the differences in structure, composition, and external properties among minerals containing illite/smectite interstratified minerals, illite, and smectite, and explored in detail the specificity of minerals containing illite/smectite interstratified minerals. It was found that illite/smectite interstratified minerals have significant differences from clay minerals such as illite and smectite in both internal nature and external properties. They are highly distinctive minerals and have excellent application potential in the cosmetics field.

2. Materials and Methods

2.1 Materials

Xuefeng mud ore was collected from the Xuefeng Mountains in Hunan Province, and the Xuefeng mud extract was obtained through processing and production by Anhui Gerui New Material Co., Ltd. Two types of illite powders for comparison were from Anhui Gerui New Material Co., Ltd. (referred to as Gerui illite) and Hebei Jinlu Biotechnology Co., Ltd. (referred to as Jinlu illite). Bentonite was purchased from Cosfad International Trade Co., Ltd.

2.2 Methods

The morphology of the samples was tested by scanning electron microscopy (SEM, Hitachi, SU8010) and high-resolution transmission electron microscopy (HRTEM, JEM-2100plus), and the crystal plane spacing of the samples was calculated. The phase structure of the samples was measured by X-ray diffraction (XRD, Bruker AXS, D8 Focus) qualitative and quantitative analysis technology and whole rock/clay analysis technology. The composition of the samples was tested by inductively coupled plasma mass spectrometry/optical emission spectrometer (ICP-MS/OES, Agilent 5110) and electron probe microanalyzer (EPMA, JEOL JXA-8530F PLUS). The contact angle was measured by a contact angle/surface tension measuring instrument (JY-82C). The viscosity was measured by a Brookfield viscometer (Brookfield, DV2T/D200), with a 2# rotor selected, a rotation speed of 200 rpm, and a test time of 40 s. The rheology was tested by a rheometer (Haake Mars40). The Zeta potential was measured by a nanoparticle size and Zeta potential analyzer (Malvern Zetasizer Nano ZS90), with a slurry water ratio of 1:10 and the pH adjusted to 9. The particle size distribution was measured by a laser particle size distribution analyzer (LS-POP-9).

3. Results and Discussion

Figure 1(a) shows the XRD results of the Xuefeng mud extract, Gerui illite, and Jinlu illite. It can be seen that all three samples contain illite. Notably, the main diffraction peak ($2\theta \approx 8.2°$) corresponding to the (002) crystal plane of the Xuefeng mud extract is significantly different from the diffraction peaks of the other two illites. The diffraction peaks on both sides of the central axis show obvious asymmetry, which is in line with the characteristics of illite/smectite interstratified minerals. In addition, the diffraction peak of the Xuefeng mud extract is short and thick, with a full width at half maximum of 0.93, much larger than that of Gerui illite (0.35) and Jinlu illite (0.22), indicating that its degree of crystallinity is low and the grains are finer.

For further analysis, whole rock/clay tests were carried out on this samples, and the results are shown in Tables 1 and 2. The Xuefeng mud extract contains 82.2% clay minerals, among the clay minerals, 73% are illite/smectite interstratified minerals, and the illite/smectite interstratified minerals accounts for 60.0%. The mixed-layer ratio is relatively low, so its crystal structure characteristics are similar to those of illite. However, Jinlu illite and Gerui illite do not contain illite/smectite interstratified minerals, indicating that the main components of the Xuefeng mud extract are different from common illites.

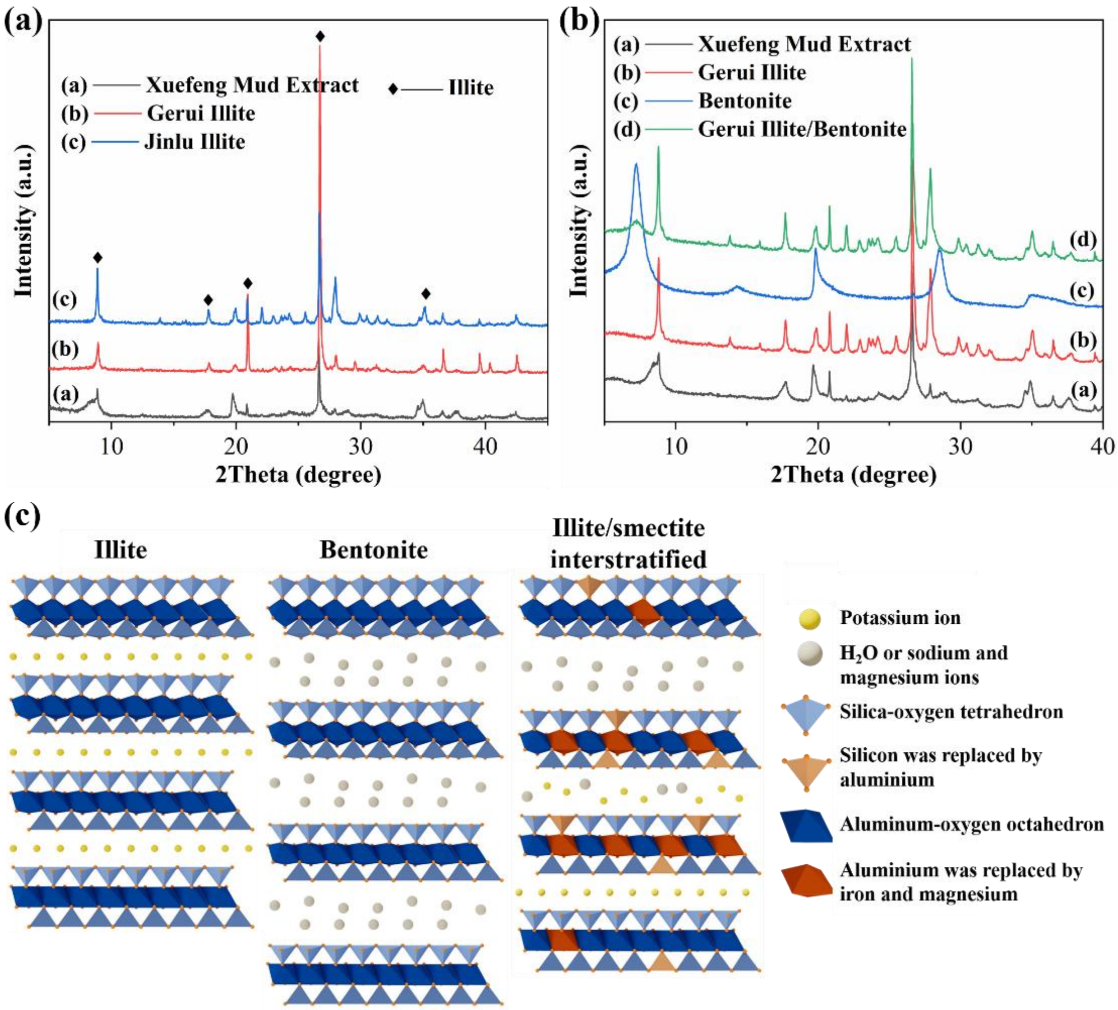

Figure 1(a) XRD patterns of Xuefeng mud extract, Gerui illite, and Jinlu illite; (b) XRD comparison of illite/smectite interstratified minerals, illite, smectite, and illite/smectite; (c) Schematic diagrams of the unit cell structures of illite/smectite interstratified minerals, illite, and smectite

To verify the essential differences in crystal structure between illite/smectite interstratified minerals and the physical mixtures of illite, smectite, and illite/smectite, in the experiment, the Xuefeng mud extract was used to represent illite/smectite interstratified minerals, Gerui illite to represent illite, bentonite to represent smectite, and the physical mixture of Gerui illite and bentonite (mass ratio, Gerui illite: bentonite = 9:1, corresponding to the illite/smectite mixed-layer ratio) to represent the physical mixture of illite/smectite. XRD results (Figure 1(b)) shown that the main component of the Xuefeng mud extract is illite/smectite interstratified minerals, and its main diffraction peak is at 2θ≈8.2°, which is significantly different from that of smectite (2θ=7.2°). The main diffraction peak of illite is thin and high, and the peak shapes on both sides of the central axis are basically symmetrical, located at 2θ=8.8°. However, the main diffraction peak of the illite/smectite interstratified minerals is short and thick, the peak shapes on both sides of the central axis are asymmetrical, and the left peak shape is severely broadened, indicating that there are significant differences in the crystal structures of the illite/smectite interstratified minerals and illite.[18] After physically mixing illite and smectite, the main diffraction peaks of illite and smectite simultaneously exist at their specific positions, while the main diffraction peak of the illite/smectite interstratified minerals does not have the main diffraction peak of smectite.

Table 1 Mineral composition and content in Xuefeng mud extract, Gerui illite, and Jinlu illite

Raw materials	**Content（%）**						
	Quartz	Potassium feldspar	Plagioclase	Calcite	Dolomite	Siderite	Clay minerals
Xuefeng mud extract	11.6	0.3	2.2	/	3.7	/	82.2
Gerui illite	64.7	1.3	5.8	2.6	4.4	1.0	20.2
Jinlu illite	21.6	1.7	17.2	/	/	2.2	57.3

Table 2 Clay mineral composition and content in Xuefeng mud extract, Gerui illite, and Jinlu illite

Raw materials	**Relative content（%）**						**Mixed-layer ratio（%S）**	
	S	I/S	It	Kao	C	C/S	I/S	C/S
Xuefeng mud extract	/	73	26	/	1	/	5	/
Gerui illite	/	/	99	1	/	/	/	/
Jinlu illite	3	/	96	1	/	/	/	/

S: Smectite group I/S: illite/smectite interstratified It: Illite K: Kaolinite C: Chlorite C/S: Chlorite/smectite interstratified

Figure 1(c) shows the schematic diagrams of the unit cell structures of the illite/smectite interstratified minerals, illite, and smectite. It can be seen that the illite/smectite interstratified minerals has a stacked structure of silica-oxygen tetrahedra and aluminum-oxygen octahedra at the micro level, with significantly different unit cell structure units compared to illite and smectite. Moreover, obvious isomorphic substitution occurs in the structure of the illite/smectite interstratified minerals compared to illite, such as the substitution of Al in the aluminum-oxygen octahedron by Fe and Mg, and the substitution of Si in the silica-oxygen tetrahedron by Al. In addition, the unit cell parameters and the contents of interlayer potassium ions and water molecules are also different, indicating that the illite/smectite interstratified minerals have very significant specificity.

Figure 2(a-c) are the SEM images of the three mineral samples. Their morphologies all exhibit lamellar structures, with the particle size of the Xuefeng mud extract being relatively smaller. This is likely due to its high degree of weathering, which reduces crystallinity and thus leads to smaller crystal grains. Figures 2(d-f) show the HRTEM images and crystal plane spacings of the three purified samples. The (002) crystal plane spacing of the Xuefeng mud extract is 1.077 nm, larger than that of Gerui illite and Jinlu illite. This is because the illite/smectite interstratified structure is in a transitional state from illite to smectite, resulting in a larger interlayer spacing, which corresponds to the lower diffraction peak angle in XRD. Figures 2(g-i) are the electron probe spectra of the three purified samples. The element concentrations of

Gerui illite and Jinlu illite are similar, while the Xuefeng mud extract contains fewer K ions, more Fe and Mg ions, and relatively fewer aluminum ions. Combining the results of ICP-MS/OES (Table 3) and XRD quantitative analysis, the chemical formula of the illite-smectite mixed-layer mineral is deduced to be $K_{0.37}(Fe,Mg)_{0.72}Al_{1.69}Si_{3.59}O_{10}(OH)_2$, which differs significantly from the common chemical formula of illite ($KAl_2Si_4O_{10}(OH)_2$). This is mainly because the Xuefeng mud extract has undergone severe weathering in a high-humidity and high-temperature environment for a long time, leading to a remarkable isomorphic substitution. Approximately 10.3% of silicon in the silica-oxygen tetrahedron is replaced by aluminum, and up to 36.0% of aluminum in the aluminum-oxygen octahedron is substituted by Fe and Mg, showing a distinct isomorphic substitution.

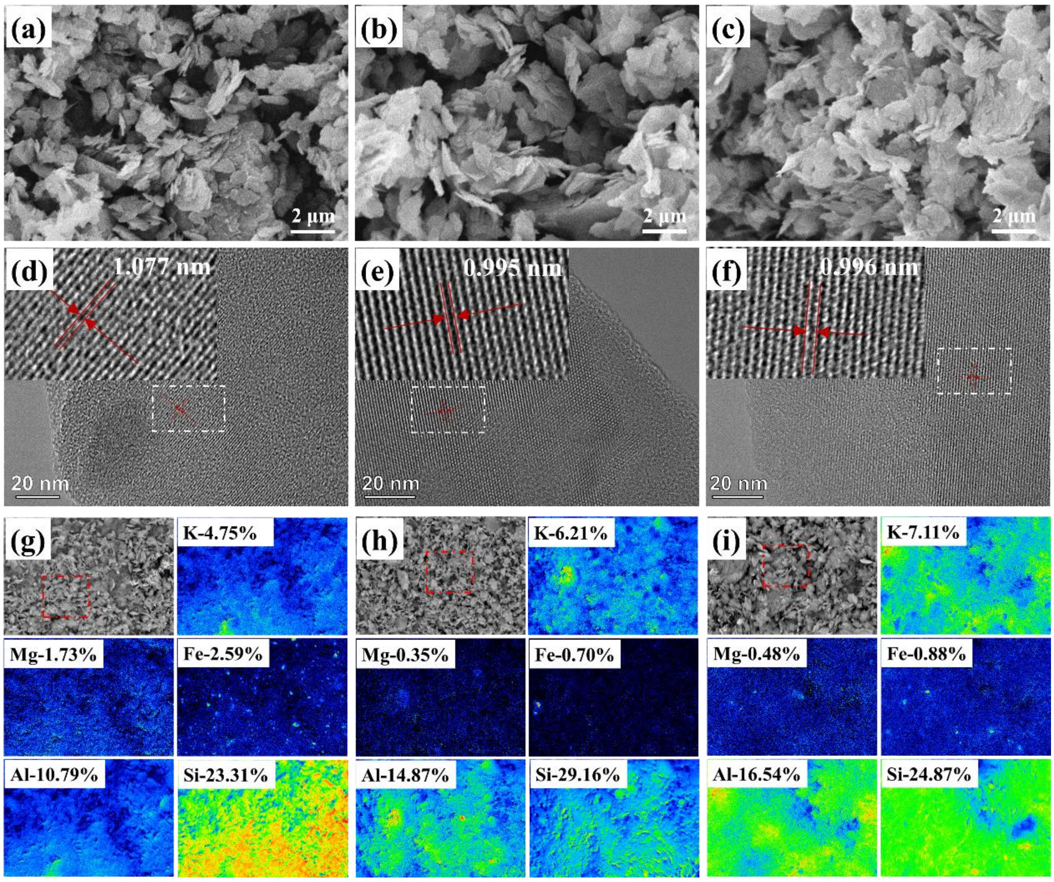

Figure 2 (a) SEM images of Xuefeng mud extract, (b) Gerui illite, (c) Jinlu illite. HRTEM images and crystal plane spacings of (d) Xuefeng mud extract, (e) Gerui illite, and (f) Jinlu illite. The element distribution and sampling point diagrams of (g) Xuefeng mud extract, (h) Gerui illite, and (i) Jinlu illite.

Table 3 Elemental Contents of Xuefeng Mud extract, Gerui illite, and Jinlu illite

Raw materials	K （mg/kg）	Mg （mg/kg）	Fe （mg/kg）	Al （mg/kg）	Si （mg/kg）
Xuefeng mud extract	9287.5	3410.1	5050.4	21205.0	45701.5
Gerui illite	11952.9	669.2	1347.5	28552.7	55838.3
Jinlu illite	13894.1	951.5	1783.0	33100.7	49823.2

Slurries with different solid-water ratios were prepared, and the viscosities of the three purified mineral raw materials were measured using a viscometer. The results are shown in Figure 3(a). When the solid-water ratio is 1:10, the viscosity values of the three slurries are low, close to that of water. At a solid-water ratio of 3:10, the viscosity of the Xuefeng mud extract slurry increases significantly (97.4 cP), much higher than that of the other two illite slurries. This is more evident at a solid-water ratio of 4:10, where the viscosity of the Xuefeng mud extract slurry (386.0 cP) is over ten times that of the illite slurry (30.0 cP). This is because the illite/smectite interstratified minerals in the Xuefeng mud extract have fewer interlayer K ions and a higher isomorphic substitution ratio, carrying more negative charges. This enhances their colloidal properties, strengthens their water adsorption capacity, reduces the free water content in the slurry, and thus increases the slurry viscosity. Additionally, the smaller particle size and high exposed layer concentration of the illite/smectite interstratified minerals enable better dispersion in water, further increasing the slurry viscosity.

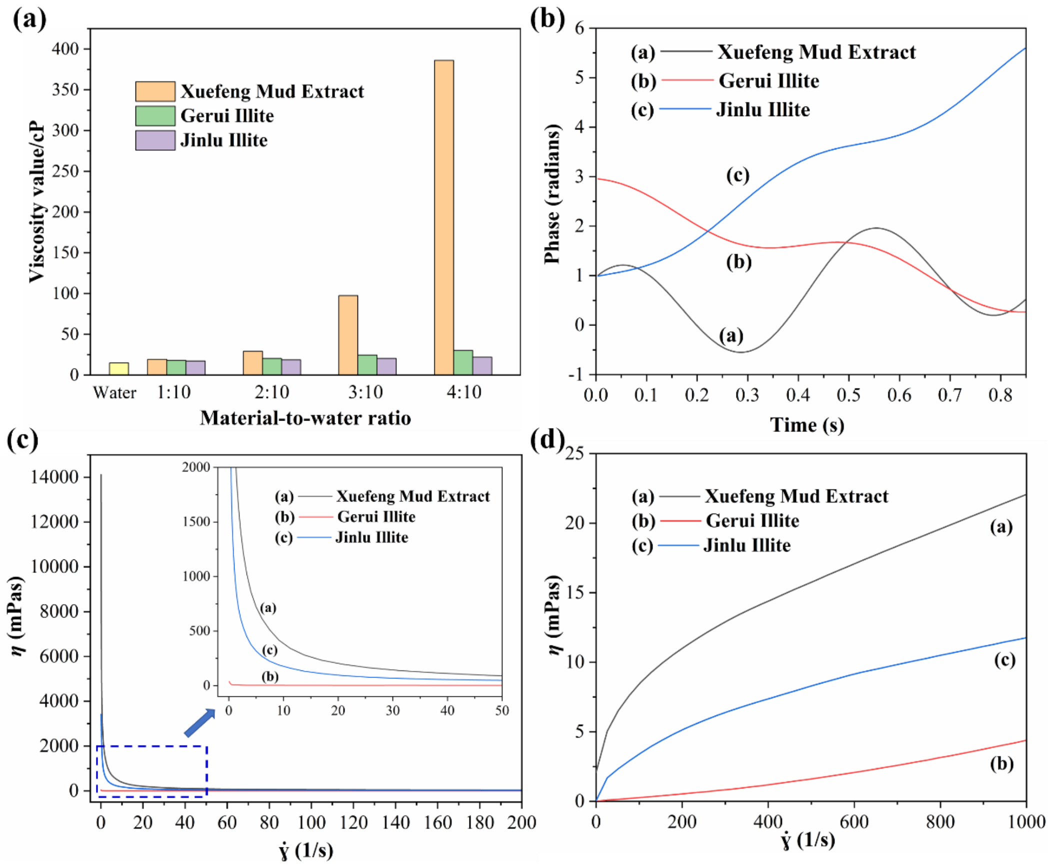

Figure 3 (a) Viscosity Spectra of Xuefeng Mud extract, Gerui illite, and Jinlu illite at Different Solid-Water Ratios; (b) Zeta Potential Diagram; Rheology Spectra of Xuefeng Mud extract, Gerui illite, and Jinlu illite: (c) Viscosity Variation with Shear Rate; (d) Shear Stress Variation with Shear Rate.

Slurries were prepared from the three purified mineral samples, and their zeta potentials were measured, as shown in Figure 3(b). The Xuefeng mud extract has a more negative Zeta potential value (-5.16 V), indicating more negative charges on its surface, more prominent colloidal properties, and better powder dispersibility. In contrast, the Zeta potential values of

Jinlu illite and Gerui illite are -1.67 V and -1.57 V respectively, with relatively low absolute values, making them more prone to agglomeration and precipitation.

The rheology of the slurries of the three purified mineral powders was tested, and the results are presented in Figures 3(c, d). Rheology refers to the property that when a fluid is subjected to an external shear force, it deforms (flows) and generates internal resistance to deformation, manifested as internal friction. Tests at different shear rates show that at lower shear rates, the viscosity of the Xuefeng mud extract slurry is much higher than that of the other two illite slurries. From the shear stress formula of non-Newtonian fluids $\eta = K\gamma^n$, the consistency coefficient K is positively correlated with the viscosity. Thus, the Xuefeng mud extract has a higher consistency coefficient, corresponding to greater internal friction of particles in the slurry. This is because the illite/smectite interstratified minerals in the Xuefeng mud extract have a stronger water adsorption force, resulting in higher consistency and viscosity at the same solid-water ratio, consistent with the viscosity test results.

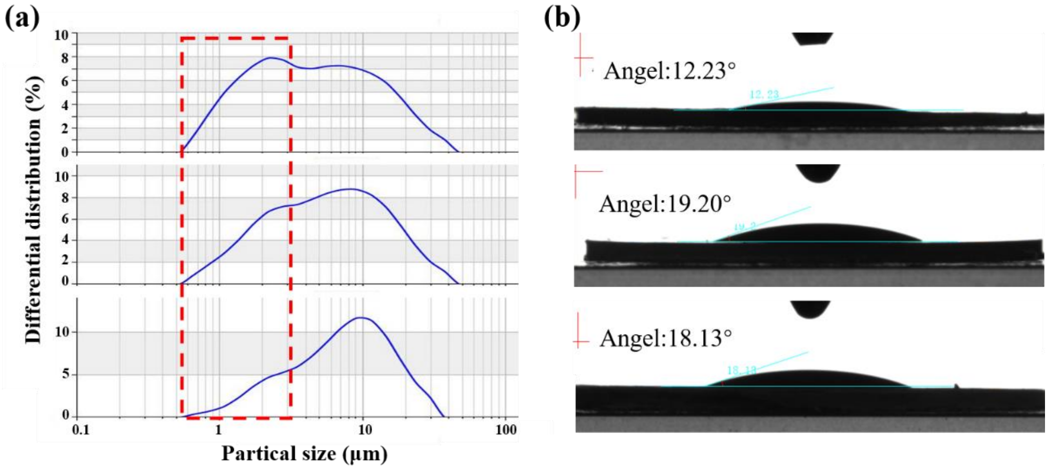

Figure 4 (a) Particle Size Distribution Diagrams of the Three Samples; (b) Oil Contact Angle Images. From top to bottom are Xuefeng Mud extract, Gerui illite, and Jinlu illite

The particle size distributions of the three mineral powders were measured using a laser particle size analyzer, and the results are shown in Figure 4(a). The Xuefeng mud extract has finer particles with a higher proportion of fine particles. This is mainly due to the high weathering degree, low crystallinity, and smaller crystal grains of its illite/smectite interstratified minerals. The three mineral powders were pressed into flat flakes, and their oil contact angles were measured using linseed oil as a reagent, as shown in Figure 4(b). The Xuefeng mud extract has a smaller oil contact angle than the other two illites, indicating a stronger affinity for linseed oil and a higher oil absorption capacity. This is because the illite/smectite interstratified minerals in the Xuefeng mud extract have more exposed surface and edge adsorption active sites, enabling better oil adsorption and resulting in a smaller oil contact angle.

Table 4 Oil Absorption Capacities of Xuefeng Mud extract, Gerui illite, and Jinlu illite

Raw materials	Oil absorption g/100g
Xuefeng mud extract	23.46
Gerui illite	21.52
Jinlu illite	21.40

The oil absorption capacities of the three mineral powders were measured using linseed oil as a reagent, and the results are shown in Table 4. The Xuefeng mud extract has a higher oil absorption capacity than the other two illites. This is because its illite/smectite interstratified minerals have smaller particles and larger crystal plane spacings, enhancing adsorption capacity, which is also reflected in the oil contact angle.

5. Conclusion

This paper conducts an in-depth and comprehensive analysis of illite/smectite interstratified minerals, and compares them with common illites on the market. A detailed exploration is carried out from the aspects of internal nature and external properties. The results show that the Xuefeng mud extract contains 60 wt.% of illite/smectite interstratified minerals, while common illite minerals on the market do not have illite/smectite interstratified minerals. The chemical formula of the illite/smectite interstratified minerals is $K_{0.37}(Fe,Mg)_{0.72}Al_{1.69}Si_{3.59}O_{10}(OH)_2$, with fewer potassium ions and more Fe and Mg ions. It is quite different from the common chemical formula of illite ($KAl_2Si_4O_{10}(OH)_2$), and has a high proportion of isomorphic substitution. Compared with common illites, the Xuefeng mud extract containing illite/smectite interstratified structures has higher viscosity, a more negative zeta potential, better rheological properties, and smaller particle size. At the same time, it has a higher adsorption capacity for oils, endowing it with better effects in terms of skin feel, fineness, refreshing feeling, and oil adsorption and control. It has excellent application prospects in cosmetics such as mud masks.

References

[1] Wang X and Wang H. Structural analysis of interstratified illite-smectite by the Rietveld method. Crystals. 2021, 28;11(3):244.

[2] Krinari GA and Khramchenkov MG. Interstratified illite–smectite phases: formation mechanisms and practical applications. Russian Geology and Geophysics. 2018, 59:1120-1128.

[3] Stixrude L and Peacor DR. First-principles study of illite–smectite and implications for clay mineral systems. Nature, 2002, 420(6912), 165-168.

[4] Deon F, van Ruitenbeek F, et al. Detection of interlayered Illite/smectite clay minerals with XRD, SEM analyses and reflectance spectroscopy. Sensors. 2022, 22(9):3602.

[5] Martín D, Aparicio P, et al. Mixed-layer illite-smectite illitization under supercritical CO2 conditions. Applied Sciences. 2022, 11;12(22):11477.

[6] McIntosh JA, Tabor NJ, et al. Mixed-layer illite-smectite in pennsylvanian-aged paleosols: Assessing sources of illitization in the illinois basin. Minerals. 2021, 11(2):108.

[7] Mills MM, Sanchez AC, et al. Understanding smectite to illite transformation at elevated (>100°C) temperature: Effects of liquid/solid ratio, interlayer cation, solution chemistry and reaction time. Chemical Geology. 2023, 615:121214.

[8] Kumari, Neeraj, et al. Basics of clay minerals and their characteristic properties. Clay Clay Miner. 2021, 24: 1-29.

[9] Bu H, Wei Y, et al. Effects of illite–smectite clay minerals on the thermal evolution of aliphatic organic matter-clay complexes: a study with thermogravimetry coupled with

Fourier transform infrared spectroscopy (TG-FTIR). Journal of Thermal Analysis and Calorimetry. 2023, 3:741-752.

[10] Christidis GE, Chryssikos GD, et al. Methods for determination of the layer charge of smectites: a critical assessment of existing approaches. Clays and Clay Minerals. 2023, 71(1):25-53.

[11] Drits VA, Sakharov BA, et al. Structural and crystal-chemical features of mixed-Layer illite-containing minerals from catagenetically altered upper jurassic oil-source rocks. Lithology and Mineral Resources. 2023, 58(3):183-95.

[12] Blanc P, Gherardi F, et al. Thermodynamics for clay minerals: Calculation tools and application to the case of illite/smectite interstratified minerals. Applied Geochemistry. 2021, 130:104986.

[13] Ghasemi M, Sharifi M. Effects of layer-charge distribution on swelling behavior of mixed-layer illite-montmorillonite clays: A molecular dynamics simulation study. Journal of molecular liquids. 2021, 335:116188.

[14] Dietel J, Ufer K, et al. Interstratified illite-hydroxy-interlayered smectite—A disorder structure model for quantification using rietveld refinement. Journal of Plant Nutrition and Soil Science. 2025.

[15] Kimura T, Kawashima H, et al. Smectite and illite/smectite mixed-layer clay minerals in the Ashibetsu coals. International journal of coal geology. 1994, 26(3-4):215-31.

[16] Inoue A, Lanson B, et al. Illite-smectite mixed-layer minerals in the hydrothermal alteration of volcanic rocks: I. One-dimensional XRD structure analysis and characterization of component layers. Clays and Clay Minerals. 2005, 53(5):423-39.

[17] Tournassat C, Gailhanou H, et al. Cation exchange selectivity coefficient values on smectite and mixed-layer illite/smectite minerals. Soil Science Society of America Journal. 2009, 73(3):928-42.

[18] Hong H, Cheng F, et al. Three-component mixed-layer illite/smectite/kaolinite (I/S/K) minerals in hydromorphic soils, south China. American Mineralogist. 2015, 100:1883-1891.