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MINERAL COMPOSITION AND COMMERCIAL APPLICATION FEASIBILITY OF SERICITE ORE IN HA TINH PROVINCE

Sericite is a silicate mineral, a finely disperse and partly hydrated variety of muscovite. It is widely applied in many areas of engineering and production, and has a high economic value. Sericite has been produced and used in the world for a few hundred years. In Vietnam, a new-discovered sericite deposit holds commercial quality reserves. This article presents the studies into the mineral composition of sericite ore from Ha Tinh deposit using a set of the modern analytical techniques, which prove efficiency of the commercial-level production and processing of this ore. The integrated studies into material constitution of Ha Tinh sericite ore show the uniform structure of sericite and insignificant content of unwanted impurities. The size of sericite particles markedly differs from the size of other minerals in the initial ore. The content of quartz gradually increases with increasing size of ore particles, while the content of sericite gradually grows with decreasing size of particles. In particularly, in very fine particles (size less than 10 μm), the content of sericite is higher than in the initial ore by 2 times and reaches 70%, while the content of quartz is 4 times less than in the initial ore. Impurities feature nonuniform distribution and occur in some mineral grains such as apatite $Ca_5(PO_4)_3$, iron oxides and binding particles of solid solutions. During ore processing, impurities can be separated to waste, and it is possible to produce a marketable product in the form of a high quality sericite powder which meets the application standards of the porcelain, ceramic, resin and other industries.

Thus, Hà Tĩnh sericite ore can be converted into valued products to be used in many various industries.

Keywords: *sericite ore, material constitution, sericite, quartz, feldspar, X-ray crystal analysis, electron microscopy, grain size composition, impurities, concentrate.*

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Introduction

Sericite represents an association of fine micaceous particles (International Mineralogical Association) [1–3]. They belong to aluminosilicate group, with a typical characteristic; the attribute of micas is their splittability to thin layers to 1 nm thick (**Fig. 1**).

The general chemical formula of this mineral group is $(K,Na,Ca)(Al,Fe,Mg)_2(Si,Al)_4O_{10}(OH)_2$. The total chemical composition of sericite is: SiO_2 43–49%, Al_2O_3 27–37%, K_2O+Na_2O 9–11%, H_2O 4–6% [4]. The chemical composition of sericite varies at different deposits depending on the general mineral composition of ore and on the content of chemical elements included in the mineral structure. The color is white, pink, light-grey or golden brown, from transparent to semitransparent, lustrous. Sometimes sericite is confused with kaolin; the difference can be sensed in touching: sericite has a

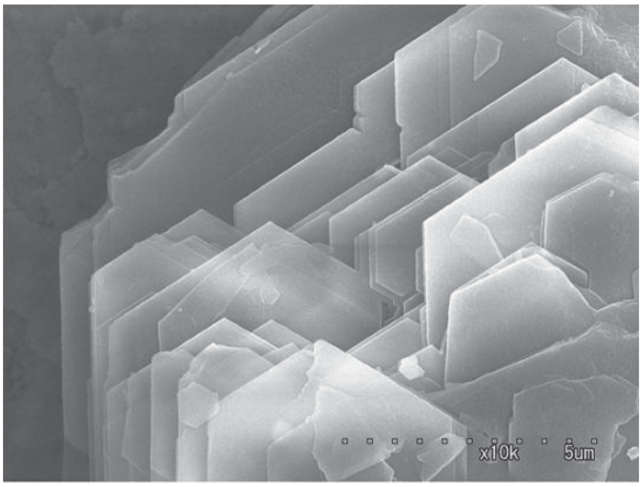

Fig. 1. Flakes of sericite under electron microscope

slippery surface, like talcum, and a characteristic silk glitter. Sericite possesses both properties of mica and clay minerals. The physical characteristics of sericite are: fine to very fine particles (usually from 4 to 30 μm); shape is from thin to very thin plates (linear dimension-to-thickness ratio is more than 80; density is ~2800 kg/m^3; Mohs hardness 2.0–3.0 [5]; high elasticity, plasticity (Young's modulus 1505–2134 MPa), smooth surface, good abrasion hardness, high temperature resistance (temperature resistance threshold 500–600 °C [6]); low thermal conductivity (heat conductivity factor 0.42–0.67 W/(m·K) [7]; specific heat 0.8 kJ/(kg·K); good insulating properties (electric strength 200 kV/mm; specific resistance 92.6 MOhm/cm) [8], high sound-proofing properties, impermeability, etc. Sericite features high chemical durability, decomposes in acid and alkaline solutions [9, 10]; well dispersive in water and organic dissolvers; has high reflectivity and refractivity; high UV radiation absorptance [9, 10]. Mining of sericite started in the middle of the 19th century. At the present time, extremely valuable characteristics of sericite have been disclosed, and, accordingly, sericite has found wider application in different industries. The demand for sericite products persistently grows worldwide.

Application of sericite in different industries

Owing to the physicochemical characteristics listed above, alongside with concentrate production at high quality and moderate cost, sericite is applied in many industries, for example as an aggregate, or a coating, or as a substance capable to expand when interacting with water or under heating [4, 8, 9, 11–13].

In the ink industry sericite is used as a swelling agent meant to enhance durability of suspensions, increase adhesion, prevent shrinkage and deformations of coatings, improve surface resistance of coating to weather, UV radiation and moisture. The characteristic silk glitter and luster make sericite irreplaceable in production of high-quality emulsion paints. For instance, cars colored with sericite-based emulsion paints have a very strong gloss and pearly luster [14].

In the paper industry sericite can be used as an aggregate in papermaking, to improve reaction sustainability and ability of paper to hold fine particles and chemicals. One of the benefits of sericite use is reduction in transparency and air permeability of paper at preserved quality of printing and at a minor decrease in reflectivity and luster [8]. Sericite can partly or completely replace kaolin [14].

In the rubber industry sericite is used as an aggregate or a mould in production of rubber goods such as tyres and roofing panels. Owing to spittability to the finest flakes, sericite acts as a lubricating and anti-adhesion agent. It can improve tension and attrition strength of rubber goods similarly to powder of carbon black (amorphous carbon) and carbon white (microcrystalline silicon dioxide). This property is of extreme value in production of white and color rubber as sericite can replace from 5 to 30% of carbon white in this case.

Production of polymers. Sericite is used as an aggregate or a swelling agent in the polymeric industry, including stiff and soft thermoplastic polymers (polyamides, polyethylene, polypropylene, polyesters, etc.) and thermosetting polymers (phenol-formaldehydes, carbamides, epoxy resins, etc.). In particular, sericite is used in production of plastics meant for acoustic and thermal insulation in cars, or as a reinforcement material in some polymeric automobile parts such as dashboards and mudguards, for improvement of mechanical strength, stiffness and part consistency.

Since the early 17th century, quartz schist–sericite is used in the *ceramic industry and in production of sanitary faience* [14, 15]. It increases heat stability, strength and decorative characteristics of ceramics.

Production of construction materials and drilling agents. In manufacturing of drilling agents, sericite can be used as an additive to bentonite. However, percentage of sericite is not higher than 1% of the mass of drilling mud heaver. Furthermore, sericite is used in production of concrete, cement-based masonry mortars, artificial paving stone, etc.

Chemical and cosmetic industry. Such properties of sericite as low hardness, microcrystalline structure (particles <10 μm), flaky shape of particles, high reflectivity, translucency and silk luster, predetermine such application field of sericite as the cosmetic industry. Considering the modern tendency of use of natural mineral products (mineral cosmetics), the price of sericite applicable in the cosmetic industry can reach USD 400–800/t.

Other fields of application. Sericite can be used in manufacture of electrodes for electro-welding, some special lubricating materials, surface coatings, coats for core pieces, materials for removal of cast from moulds in metallurgy, UV radiation filters, etc. Moreover, sericite is used in electronic engineering, in production of composite plastics, insulating tape for electrical insulation (mica tape), etc.

Material constitution of sericite ore from Ha Tinh Province

The leading countries producing and using sericite are the USA, Russia, South Korea, Canada, France, Taiwan, Malaysia, Brazil, Mexico, India and Sri Lanka. Some of the top producers of sericite are Shanshinsericite, Myoshi Kasei, Nikko Toryo (Japan), CAS (South Korea), Chuzhou Grea Mineral, Mitsui China (China) and other. Sericite production in 2017 totaled 356.000 t, including 100.000 t in China, 57.900 t in Finland, 40.000 t in the USA, 24.000 t in Canada, 21.000 t in France and 4.000 t in India [16]. By estimates of the U.S. Geological Service, the demand for sericite products, mainly powders, will grow by 1–3% yearly. The key application fields are production of powder for manufacture of binders and filler

Fig. 2. Ha Tinh Province, Vietnam

Fig. 3. Ha Tinh sericite ore

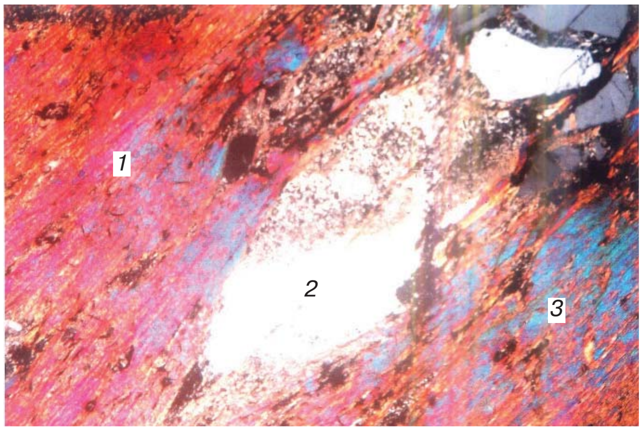

Fig. 4. Microsize flakes of sericite (1) in parallel strips interlaced with quartz (2) and feldspar (3); 25-fold augmentation, Nikon ECLIPSE LV100-POL

pasts in the construction industry, manufacture of coatings and polymers, special plastics for the automobile industry, and feedstock for the cosmetic industry.

Vietnam launched sericite exploration in the 1990s. Sericite was discovered in the provinces of Quang Tri, Quang Ninh, Bac Kan, Kon Tum and Ha Tinh (**Fig. 2**). At the same time, sericite content of most ore bodies in these provinces is low. The discovered ore bodies are small, sericite is of low grade, and the resources have no commercial value. The exclusion is Ha Tinh Province [17]. According to geological exploration and appraisal accomplished by the North Central Geological Division, Ha Tinh sericite is concentrated in some ore bodies suitable for commercial operation and sericite production as sericite reserves make a few million tons (**Fig. 3**).

The material constitution of Ha Tinh sericite ore was determined using:

— X-ray crystal analysis and diffractometer D8-Advance;

— differential thermal analysis and thermoanalyzer STA-PT 1600;

— micro zone analysis and electron microscope Camebax,

— analysis on inductively coupled plasma mass spectrometer Iris-Intrepid;

— atomic absorption analysis.

The studies were carried out at the Center for Analytical Services of Experimental Geology at the Institute of Environmental Technology under the auspices of the Vietnam Academy of Science and Technology. Mineralogical and petrographic analyses were implemented using polarizing microscope AXIOLAB at the Institute of Geosciences and Mineral Resources. The structural analysis was executed on a scanning electron microscope at the Institute of Material Sciences, VAST, and on a transmission electron microscope at the Institute for Geography and Geology, University of Greifswald in Germany.

The research shows a sufficiently high content of sericite in crude ore samples from Ha Tinh. The content of impurities—e.g. iron oxides, is low but metallic impurities are associated with numerous nonmetallic impurities, such as quarts, feldspar, etc. Some of the research findings are presented below in this paper.

Thus, Ha Tinh ore without processing is off the quality standards for production of aggregates for the pink, polymeric and, particularly, cosmetic industries.

The X-ray crystal analysis shows that sericite ore includes such prime minerals as:

— sericite, clearly distinct at peaks with an angle 2θ, equaling 10.3°; 20.8°; 31°; 32.5°; 41.5°; 49.3° and 53°; average composition $(K_{0,727}Na_{0,17}Ca_{0,011})Al_{0,933}Fe_{0,016}Mg_{0,011})_2(Si_{0,782}Al_{0,221})_4O_{10}(OH)_2.5H_2O$;

— quartz (SiO_2);

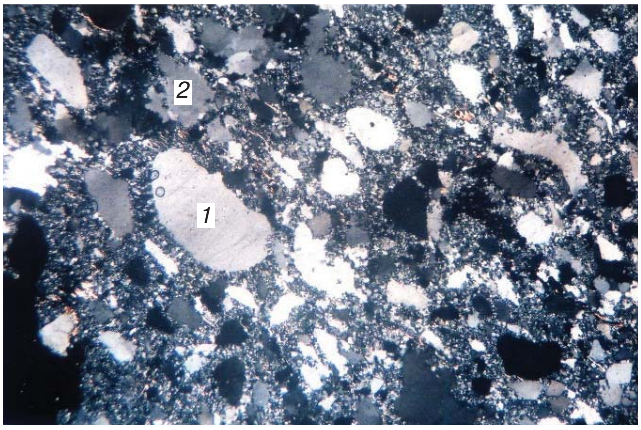

Fig. 5. Prismatic quartz crystals with rounded edges (1), eroded feldspar (2) and sericite aggregates (3); 25-fold augmentation, Nikon ECLIPSE LV100-POL

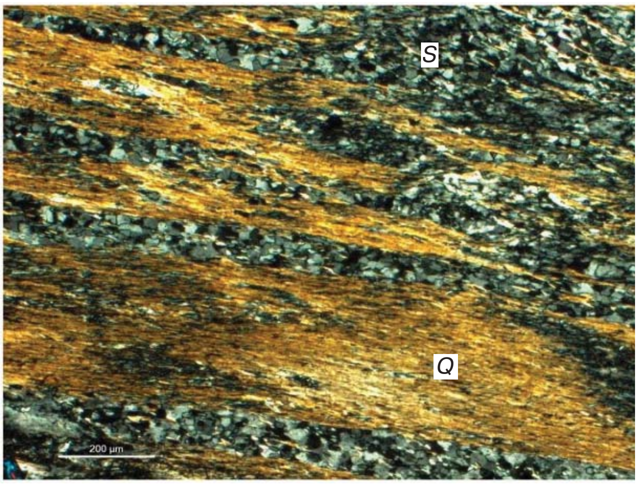

Fig. 6. Tightly associated particles of sericite *S* and quartz *Q*

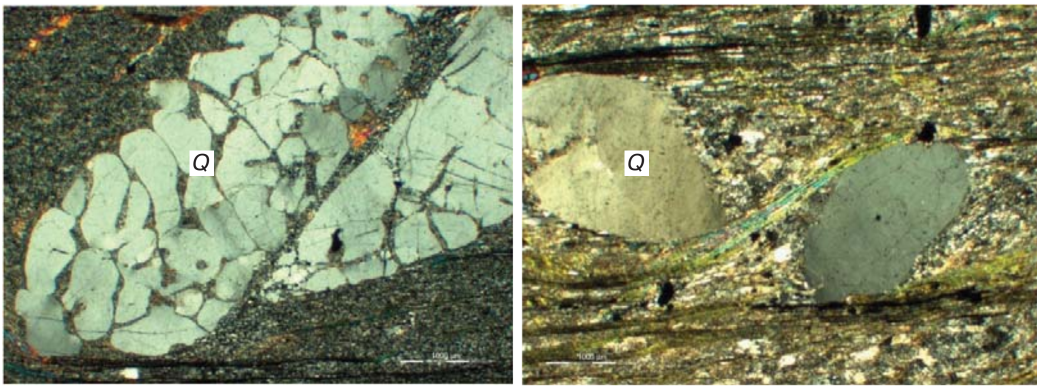

Fig. 7. Particles of quartz *Q* in sericite *S*

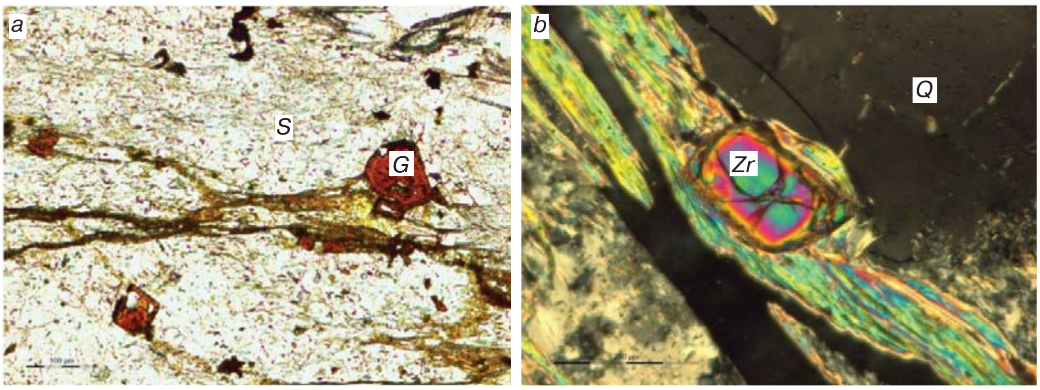

Fig. 8. Particles of hematite G in sericite S (*a*) and particles of zircon Zr and sericite S in quartz Q and feldspar Fs

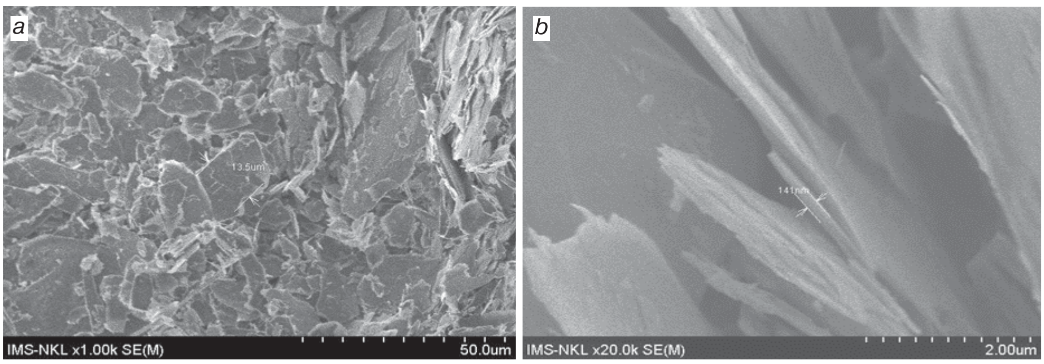

Fig. 9. Image of Ha Tinh sericite sample on SEM at augmentation of x1000 (*a*) and x20000 (*b*)

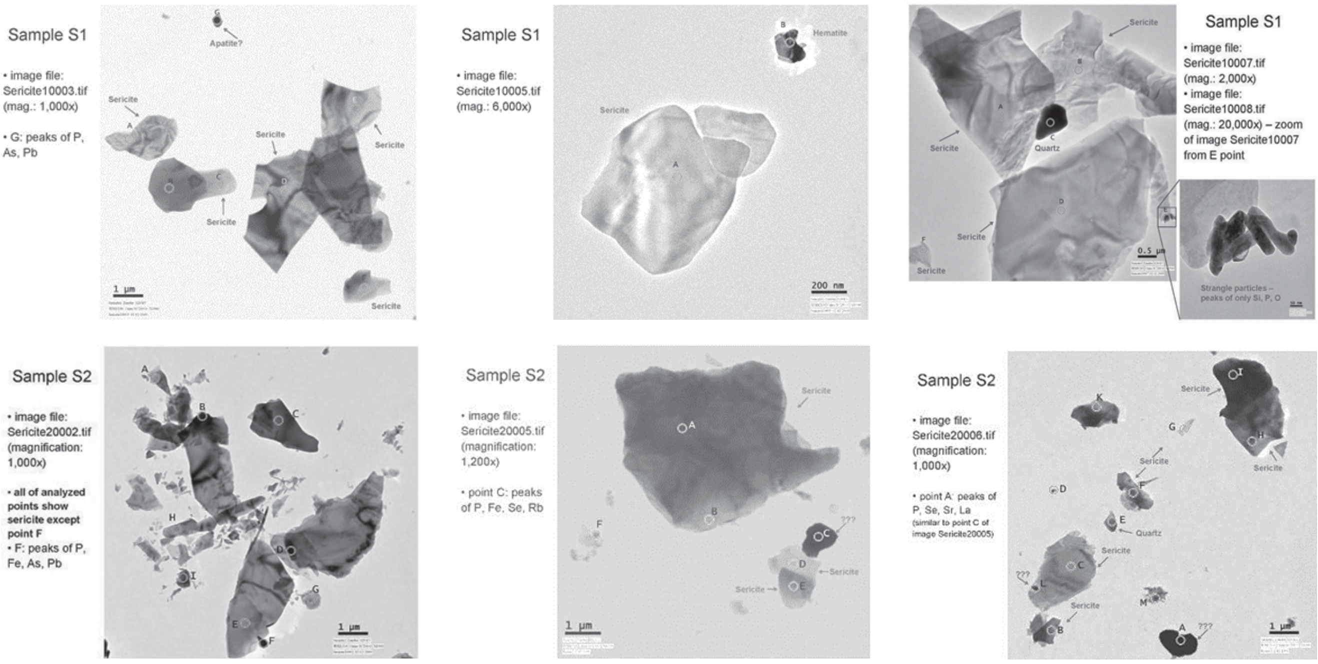

Fig. 10. Images of Ha Tinh sericite samples on transmission electron microscope

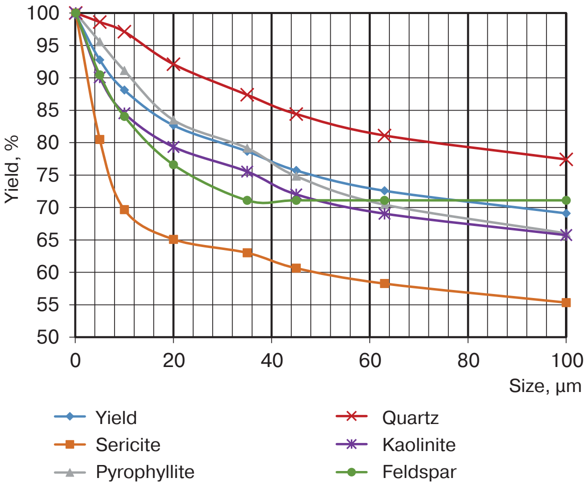

Fig. 11. Granulometry of sericite ore and mineral distribution in size grades

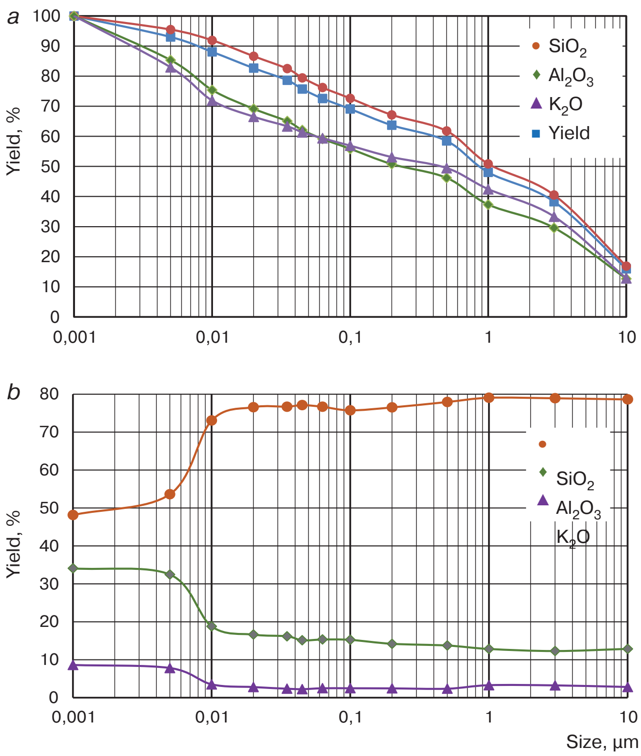

Fig. 12. Distribution (*a*) and content (*b*) of basic elements in different size grades of Ha Tinh sericite ore

— pyrophyllite $Al(Si_2O_5(OH)$, distinct at peaks at an angle 2θ, equaling 11.3°; 19.5°; 23.5°; 24°; 29°; 31°; 34°; 46.2°; 49.5° and 53.8°;

—kaolinite $Al_2SiO_5(OH)_4$;

—chlorite;

—low albite–oligoclase with average formula $(Na_{0,75}Ca_{0,25})(Al_{1,26}Si_{2,74}O_8)$.

The mineralogical and petrographic analyses show that 97–99% of crude ore from Ha Tinh Province is composed of quartz, sericite and feldspar $KAlSi_3O_8$ in the form of very tight accretion of micron particles (**Figs. 4 and 5**). Sericite particles have sizes from a few hundred nanometers to a few hundred micrometers (**Fig. 6**). Quartz crystals have usually shapes of prisms with round edges and convex or concave faces, up to 0.2 to 0.3 mm in size (See Fig. 5). Quartz can occur as microparticles or nests in particles of other minerals, having the least size of 30 µm. Pyrophyllite grains from 3 to 20 µm in size contain many hidden flakes arranged in parallel to sericite aggregates. The content of pyrophyllite varies from 2 to 65%. Feldspar particles have either tabular or distorted

Table 1. Chemical composition of sericite samples from Hà Tĩnh deposit

Sample	Content, %						
	SiO_2	Al_2O_3	MgO	K_2O	FeO	MnO	H_2O
M-1	45.56	37.60	0.27	10.80	0.33	0.11	4.60
M-2	45.52	36.83	0.16	10.60	0.45	0.14	5.01
M-3	45.86	37.56	0.20	11.28	0.21	0.16	4.65
M-4	46.12	36.28	0.13	10.98	0.22	0.28	5.12
M-5	46.02	37.06	0.08	11.35	0.34	0.22	4.51

Table 2. Grain size analysis of sericite samples

Sample	Yield (%) of different sizes (μm)					
	–10+0	–20+10	–35+20	–63+35	–100+63	+100
M-1	50.83	13.05	12.75	1.45	0.22	21.7
M-2	56.99	10.8	10.65	1.91	0.83	18.82
M-3	58.54	10.52	8.5	1.61	0.7	20.13
M-4	26.58	14.56	32.7	8.09	1.87	16.2
M-5	34.2	15.35	26.95	4.03	0.95	18.52

shapes and sizes from 0.5 to 3 mm (See Figs. 4 and 5). Micron particles (maximum size of 30 μm) are seen in other minerals. The content of feldspar is 50–60% in unaltered rocks, 20–30% in weakly altered rocks and not more than 3–5% in heavily altered rocks interlaced with quartz (Fig. 5).

The cementing agent is lava ash transformed as a result of weathering into a conglomerate of fine sericite and clay flakes interlaced with extremely fine quartz. Only 1–2% of quartz crystals are fissured rounded particles 0.5–2.5 mm in size (**Fig. 7**). Sometimes acidic volcanic rocks (porphyritic quartz) with particles of 4 mm are observed.

Furthermore, the ore contains to 1–2% of some metallic minerals. Some ferrous minerals (hematite, goethite, limonite, iron hydroxides, pyrite) are mostly uniformly distributed in the ore as fine flakes less than 10–100 μm (**Fig. 8a**).Chalcopyrite, ilmenite, sphene and zircon are observed sometimes (**Fig. 8b**). Titanium-bearing minerals, such as leucoxene, rutile and sphene, have sizes of 100–500 μm and are disseminated in the nonmetallic matrix. Sulphide particles have an average size less than 10 μm and are as a rule nested in some nonmetallic minerals.

Purity of Ha Tinh sericite was analyzed on electron microanalyzer Camebax (**Fig. 10**), which, in combination with the chemical analysis (**Table 1**) has allowed finding that:

—the samples mostly have the same chemical composition;

—Ha Tinh sericite features high purity, is relatively uniform and contains more than 10% of K_2O, which approximately equals the theoretical content of 9–11%;

—the content of ferrous impurities is very low.

According to the grain size analysis data (**Table 2**), sericite is mostly represented by particles less than 20 μm (size yield is more than 60%), i.e. sericite is sufficiently fine to meet application standards in many fields of use.

The chemical analysis of Ha Tinh sericite using the inductively coupled plasma method shows (**Table 3**) that the test sample contains 16.20% of Al_2O_3, 3.52% of K_2O 3.52% and relatively low iron hydroxide Fe_2O_3 (0.23%). The content of

Table 3. ICP-based chemical analysis

Oxide	Content, ‰	Element	Content, ‰	Element	Content, ‰	Element	Content, ‰
Al_2O_3	16.20	Ag	< 2	Cu	< 5	Sc	9.11
CaO	0.074	As	31.66	Ga	20.03	Sn	10.75
Fe_2O_3	0.23	B	< 10	Ge	< 20	Sr	29.23
K_2O	3.52	Ba	358.55	La	363.23	Ta	< 10
MgO	0.024	Be	< 5	Li	< 5	V	40.71
MnO	< 0.05	Bi	< 10	Mo	< 5	W	< 20
P_2O_5	0.005	Cd	< 2	Nb	13.78	Y	52.11
TiO_2	0.63	Ce	117.65	Ni	12.11	Zn	14.33
SiO_2	74.48	Co	2.40	Pb	36.82		
		Cr	17.59	Sb	< 10		

Table 4. Content of basic minerals in different size grades of Hà Tĩnh sericite ore

Size grade, μm	Yield, %	Content, %				
		Sericite	Pyrophyllite	Quartz	Kaolinite	Feldspar
+ 100	69.10	20	15	56	4	3
–100 + 63	3.48	21	20	53	4	-
–63 + 45	3.13	19	22	53	4	-
–45 + 35	2.96	20	23	50	5	-
–35 + 20	4.00	13	17	59	4	4
–20 + 10	5.44	21	22	46	4	4
–10 + 5	4.66	58	15	16	5	4
–5 + 0	6.96	70	10	10	6	4
Total	100.0	25.14	15.17	50.02	4.29	2.93

some heavy metals is at the level of units or tens (seldom hundreds) per mill, for instance, arsenic content is 31.66‰ and lead content is 36.82‰.

The transmission electron microscopic analysis revealed heavy metal impurities (As and Pb) in some minerals such as apatite $Ca_5(PO4)_3$, iron hydroxides or ore particles in the form of solid solutions (Fig. 8b).

The grain size classification was implemented using the screen analysis for large particles and sedimentation for fines. The data of the material analysis (**Table 4**, Fig. 11) and chemical analysis of sericite ore, as well as size grading (**Fig. 12**) show that the ratio of their contents noticeably change subject to particle size. The content of quartz gradually increases with increasing size, while the contents of K_2O and Al_2O_3 increase with decreasing size of particles. Size grade −10 µ, contains more than 8.3% of K_2O and more than 34% of Al_2O_3, which is 2 times higher than in the initial ore. The content of quartz in this size grade is less than 25% than in the initial ore. The distribution of K_2O and Al_2O_3 in this size grade is 25 and 28%, respectively.

Conclusions

The integrated studies into material constitution of sericite ore from Ha Tinh deposit have allowed the following conclusions to be drawn:

1. The composition of minerals in the initial ore samples is relatively uniform. Minerals are represented by particles from a few hundred nanometers to a few hundred microns in size, bound by lava ash transformed into fine disperse sericite and clay flakes interlaced with micron particles of quartz.

2. The size of sericite particles markedly differs from the size of other mineral impurities in the initial ore samples. The content of quartz gradually grows with increasing size of the particles, and the size of sericite gradually increases with increasing size of the particles. In particular, very small grains (less than 10 µm) contain 2 times more sericite (up to 70%) than the initial ore sample, while the content of quartz is 4 times less than in initial ore.

3. The content of impurities including iron and heavy metals such as arsenic and lead is higher than the standard content for commercial production of sericite powder for the chemical and cosmetic industries.

4. The impurities are uniformly distributed and occur in grains of some minerals such as apatite $Ca_5(PO4)_3$, hydroxides and binding particles of solid solutions. Consequently, during processing, they can be separated into dump waste product toward marketable production of sericite powder.

5. Based on the features of the composition and value of initial sericite ore from Ha Tinh deposit, the processing methods can be:

— selective crushing and / or milling to dissociate sericite particles and gangue particles;

— hydraulic classification of sericite particles less than 10 µm in size;

— flotation of sericite particles more than 10 µm size, separation of these particles from quartz and other mineral impurities, and milling down to the size required for the ink and polymer industries;

— chemical treatment of sericite flotation concentrate in order to remove impurities of iron and heavy metals, and to produce sericite powder suitable for the chemical and cosmetic industries;

— development of a technology to remove impurities of iron and heavy metals toward obtaining high quality products as per the standards of the specific industries such as porcelain, ceramic and resin industries, etc.

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