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The influence of process parameters on the structure and properties of micamechanically activated in an ultra centrifugal mill

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This paper presents the results of research on mechanical activation of dry mica in an ultra centrifugal mill with a peripheral comminution path.
The variable parameters of the mill operation were: rate of rotor revolution (n_o = 10.000 and n_o = 20.000 r.p.m.), circle sieve mesh (80, 120, 200 and 500 μm) and the current intensity. The following technological parameters were studied: mechanical activation time, rotor velocity, mill capacity and specific energy consumption. The mechanically activated powder was examined by application of thermal and thermogravimetric analyses, analysis of the degree of mechanical activation and the specific surface area, as well as X-ray diffraction analysis.
The optimal results of the mechanical activation were obtained with a full mill load, using a circle sieve mesh of 80 μm and nominal rate of rotor revolution of 20.000 r.p.m. It was shown that the mechanically activated mica obtained employing these process parameters had passed into the amorphous state.

Key words: Mica, Amorphous materials, Ceramics, Thermogravimetric analysis (TGA), Crystal structure.

Introduction

The quality requirements of mechanically activated mica are high for its application in the synthesis of contemporary materials. Due to their physical properties, micas have significant importance both, in conventional applications such as in plastics, pearlescent pigments, optical filters, stove windows, condensers, rheostats, fuses, insulators, drilling muds, adsorbents, fire extinguishers, concretes, etc. and in nanotechnology, where they are extensively used as a substrate for deposition or self-assembly, as templates in the preparation of nanowires and in clay–polymer nanocomposites [1].

Mica glass-ceramics are widely used as mechanical, electrical, and biomedical materials due to their unique machinability and good electrical properties. By using a novel hot-pressing technique, it is possible for a new type of mica glass-ceramics to be prepared where the microstructure consists of preferentially aligned mica particles [2].

For many applications, it is necessary to reduce the particle size of the natural raw materials. An alternative route for dry mechanical activation of minerals of mica using knife-mills was found. Comminution by cutting was shown as the most efficient technique to produce material with a size below 100 mm without morphological or structural alterations [3].

Grinding, either in the dry state or in the presence of water and chemical additives, is the common procedure used for particle size reduction of these materials. Different grinding procedures are used in industry for micas: dry grinding (yielding particles in the range from 1.2 mm to 150 m), wet grinding (95-45 μm), or mechanical activation (< 53 μm) [4]. It is well known that grinding not only reduces the particle size but also has various effects on the structure and laminar silicates properties, such as amorphization, aggregation or modification of the surface properties that are undesirable in general. In addition, particle size reduction of clay and mica minerals by a high-energy ball milling (HEBM) technique has been widely studied. Particle size reduction of clay and mica by a HEBM technique has a strong influence on their surface physical-chemical characteristics, i.e., the specific surface area (SSA), the cation exchange capacity (CEC), and the electrokinetic properties. Significant changes of the size, morphology and structure were followed by the change of the physical-chemical properties [5].

The mica powder should consist of a fine particle size distribution, with strictly defined physical-mechanical, physical-chemical and mineralogical characteristics. Every deviation from the required characteristics exacerbates the structure and properties of mechanically activated products. Under fine and ultra-fine grinding,

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the mechano-chemical alterations in the structure of the particles of the material occur initially in the surface layer. The rate of alteration in the initial phases of mechanical activation is determined by the rate of increase in the specific surface area, which depends on the physical properties of the material and the possibilities of the apparatus employed for mechanical activation to efficiently transfer a major amount of the mechanical energy onto the material [6-9].

The process of mechanical activation of mineral raw materials, including micas, is not a simple process and has been studied widely during the last few decades. Two methods of mechanical activation (ball milling and oscillatory milling) have been studied intensively. Mechanical activation using a ball mill was found to be a slow process where dry grinding proved to have some effect compared to wet grinding. In addition, mechanical activation in an oscillatory mill could produce almost complete destruction. These findings referred to clay minerals [10-14].

The results of the kinetics and mechanisms of the mechanical activation process of these minerals in a centrifugal mill presented in this paper represent a contribution to the investigation and determination of the occurrences and processes occurring in the material under the effects of mechanical forces during dispersion. The changes in the structure and properties, as well as of the energy state of the materials, were studied. Variations in the parameters of the grinding process were realized with the goal of optimization and automation of the grinding processes in a centrifugal mill. The results obtained served as a base for determining the correlation of the parameters of the grinding technology with the structure and desired properties of the resulting mica powders, as well as for forecasting their application in the synthesis of contemporary materials.

Experimental

The investigations of the mechanical activation of dry micas were realized in five series of experiments. As the initial material, a flotation mica concentrate obtained by the technological processing of white granites from the bed "Samoljica", Bujanovac, Serbia, was used, the series designated M_0. The grindings were realized in a centrifugal mill with a peripheral comminution path. The mill could be operated at nominal rotor speeds of n_0 = 10.000 and n_0 = 20.000 r.p.m. In addition, it was possible to choose a circle sieve of different mesh sizes (80, 120, 200 and 500 μm) and vary the intensity of the current. The series performed with the mesh sizes 80, 120, 200 and 500 were designated M1, M_2, M_3 and M_4, respectively.

The process of kinetics was analyzed through the changes in the particle size distribution, respectively the specific surface area, with the time of mechanical activation. The analyses of the mechanical activation products were realized using different instrumental techniques. For detailed powder characterization (determination the particle size distribution, the mean diameter and specific surface area of the grains) a "Coulter-Electronics-Coulter Multisizer", which possesses software for all important correction factors for determination of the physical characteristics of powders, was used. To define the thermal and thermogravimetric characteristics of the samples, a simultaneous thermal analyzer "STA-409 EP" was used. X-Ray structural analysis was utilized for the determination and observation of the phase composition of the refractory drivers. For this

Table 1. The characteristics of the initial mica sample (M_0)

Particle size distribution				Mineralogical composition		Chemical composition	
Size class (mm)	Mass M (%)	Cumu. Over. R (%)	Cumu. under. D (%)	Mineral	Cont. Wt%	Comp.	Content, Wt%
							Size class – 0.589 + 0.104 mm
-0.833 + 0.589	0.10	0.10	100.00			SiO_2	56.60
-0.589 + 0.417	4.40	4.50	99.90	K-muskovite	74.11	Al_2O_3	24.50
-0.417 + 0.295	22.50	27.00	95.50			CaO	0.10
-0.295 + 0.208	29.00	56.00	73.00			MgO	0.60
-0.208 + 0.147	23.00	79.00	44.00	Na-muskovite	10.13	Na_2O	1.90
-0.147 + 0.104	10.50	89.50	21.00			K_2O	10.59
-0.104 + 0.074	6.00	95.50	10.50	Quarz	7.19	Fe_2O_3	1.00
-0.074 + 0.063	1.40	96.90	4.50			MnO	0.03
-0.063 + 0.053	1.10	98.00	3.10			TiO_2	0.18
-0.053 + 0.040	0.82	98.82	2.00	Feldspar	3.74	P	0.40
-0.040 + 0.000	1.18	100.00	1.18			S	0.40
Total	100.00	-	-	Total	95.17	I.L.	3.70

Table 2. Process parameters related to the ultra centrifugal mill

Series	Test No.	Parameters related to the ultra centrifugal mill			
		Rotor revolutions (r.p.m.)	Actual rot. rev. (r.p.m.)	Sieve mesh (μm)	Intensity of current (A)
M_1	M_{11}	10,000	10,824.26	80	2.30
	M_{12}	20,000	19,314.29	80	2.80
M_2	M_{21}	10,000	13,360.13	120	1.60
	M_{22}	20,000	20,626.48	120	2.50
M_3	M_{31}	10,000	14,435.96	200	1.40
	M_{32}	20,000	22,213.92	200	3.20
M_4	M_{41}	10,000	11,738.24	500	2.80
	M_{41}	10,000	11,738.24	500	2.80

Table 3. Technological parameters of mechanical activation

Series	Test No.	Technological parameters of mechanical activation			
		Process duration (min)	Rotor speed (m/s)	Mill capacity (kg/h)	Specific energy consumption (kWh/t)
M_1	M_{11}	30.00	74.40	0.150	3080.00
	M_{12}	12.00	100.24	0.375	1642.66
M_2	M_{21}	6.00	74.40	0.500	616.00
	M_{22}	6.00	114.70	0.500	968.00
M_3	M_{31}	4.50	74.40	1.000	461.77
	M_{32}	6.00	114.70	0.500	968.00
M_4	M_{41}	4.00	81.63	0.750	352.00
	M_{42}	4.00	114.70	0.750	645.33

purpose, an X-ray diffractometer "PW-1710" was used.

The results of the particle size distribution, chemical and minerological composition of the initial sample (M_0) are given in Table 1.

During all experiments in all series, the process parameters of dry mechanical activation were observed, *i.e.*, mechanical activation time (t), rotor speed (v), mill capacity (Q) and the specific energy consumption (e).

For the characterization of the mechanical activation products, the following relevant parameters were studied: (d_1) and (d_2) - the sieve mesh sizes through which the samples were passed through (μm); (R_1) and (R_2)- the cumulative screen outs, (%); the d' parameter- depends on the particle size distribution of the sample, which gives information concerning the massiveness of a sample and represents the sieve mesh size where $R = 36.79\%$; n- a direction coefficient which depends on the particle size distribution of the sample; d_{95}- the sieve mesh through which 95% of the mechanically activated product passes (μm); S_1- theoretical specific surface area (m^2/kg); S_r- the real specific surface area (m^2/kg). The results of the measurements of the process parameters related to the mill operation and the parameters connected to the mechanical activation process are given in Tables 2 and 3, respectively.

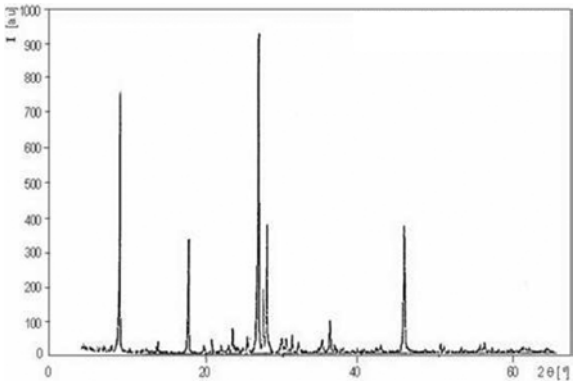

Fig. 1. Diffractogram of the initial mica sample (M_0)

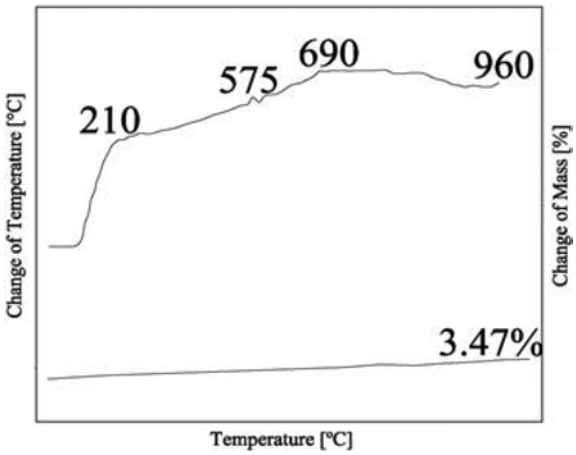

Fig. 2. DTA and TGA curves of the initial mica sample (M_0)

Results and discussion

The results of the X-ray and thermal analysis of the initial mica sample, series (M_0), are shown in Figs. 1 and 2, respectively.

In accordance with the data obtained by chemical analysis (Table 1.), it can be seen in Fig. 1 that the initial sample (M_0) was crystalline mica (muscovite), which was also registered by the endothermic effect at 575 °C in Fig. 2. In addition, in the same Figure, the TGA results show that the sample gradually lost mass on heating to 1000 °C ; the total mass loss being 3.47%.

The values of the parameters measured to observe the mechanical activation process and evaluate the quality of the mechanical activation products in series M_1, M_2, M_3 and M_4 are given in Table 4.

It was noted that the mechanical activation rate, as the main characteristic of mechanical activation kinetics, increases with an increase in the circle sieve mesh from 80 to 500 μm and with an increase in the mill load. The rate maximum is at the nominal mill load and at the highest rate of mill rotor revolutions, 20.000 r.p.m.

According to the results of the particle size distribution

Table 4. Parameters of the mechanical activation products

Series	Test No.	Parameters of the mechanical activation products								
		d_1 /μm/	d_2 /μm/	R_1 /%/	R_2 /%/	δ /μm/	n	d_{95} /μm/	S_t /$m^2\,kg^{-1}$	S_r /$m^2\,kg^{-1}$
M_1	M_{11}	5.00	40.00	92.66	3.60	20.55	1.79	37.20	181.38	544.14
	M_{11}	5.00	30.00	90.02	5.10	16.40	1.86	28.94	218.19	654.57
M_2	M_{21}	5.00	63.00	95.70	4.00	27.47	1.80	49.25	134.92	404.76
	M_{22}	5.00	50.00	94.00	5.00	21.99	1.84	38.93	164.55	493.65
M_3	M_{31}	5.00	63.00	95.20	4.20	25.50	1.82	45.42	143.60	430.80
	M_{32}	5.00	69.00	94.00	0.90	22.48	1.82	40.04	162.89	488.67
M_4	M_{41}	5.00	147.00	99.43	4.00	79.15	1.86	139.29	45.21	135.63
	M_{42}	5.00	147.00	99.36	13.00	71.31	1.88	124.74	49.63	148.89

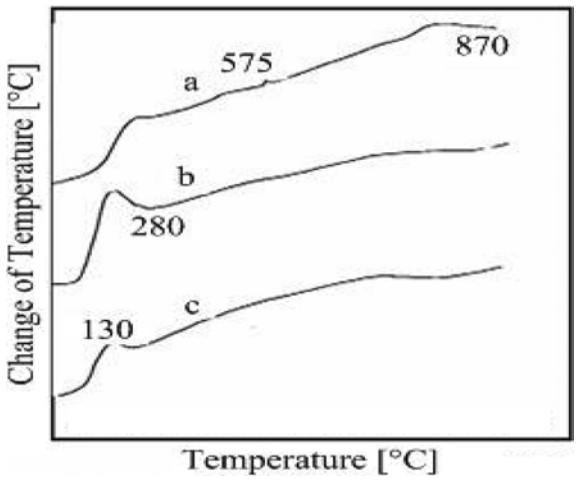

Fig. 3. Comparative DTA results for (M_0), b) (M_{11}), c) (M_{12})

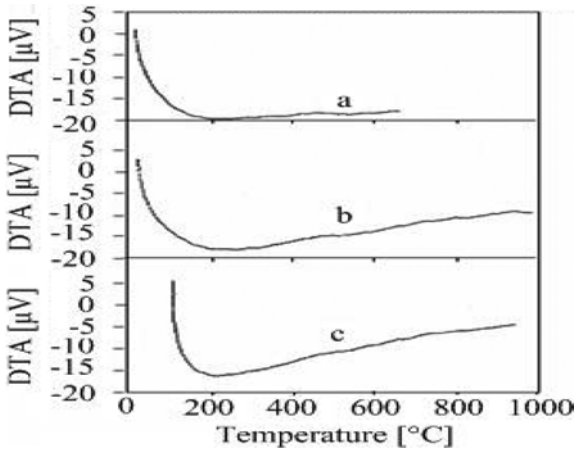

Fig. 4. Comparative TGA results for samples (M_0), b) (M_{11}), c) (M_{12})

analysis (Table 4.), it can be observed that changing the circle sieve mesh from 80 to 500 μm and decreasing the nominal rate of rotor revolution from 20.000 r.p.m. to 10.000 r.p.m. led to increased massiveness in the particles of the mechanically activated product. The finest product in terms of its massiveness was obtained with a circle sieve mesh of 80 μm and a nominal rate of rotor revolution of 20.000 r.p.m. Therefore, it could be concluded that with an increase in the circle sieve mesh, the mechanical activation rate, as the main characteristic of mechanical activation kinetics, also increases. Basically, the circle sieve mesh is not directly connected to mechanical activation kinetics, it is more related to fineness; actually, the desired fineness of mechanical activation dictates which circle sieve and which mesh should be used.

For a detailed structure and properties analysis of mechanically activated mica, samples obtained in the ultra centrifugal mill with a sieve mesh of 80 μm at a nominal rate of mill rotor revolutions of 20.000 r.p.m. were used, respectively 10.000 r.p.m., samples of the series (M_1). Comparative DTA and TGA results are shown in Figs. 3 and 4 for: a) the initial sample, (M_0), b) a mica sample mechanically activated at a nominal rotor rate of 10.000 r.p.m., (M_{11}) and c) a mica sample mechanically activated at a nominal rotor rate of 20.000 r.p.m. (M_{12}).

It can be seen from the comparative diagrams that, at the beginning of heating up to 200 °C, respectively 250 °C, a change in the baseline of the curves of all samples occurred, by even up to 20 μV. This change was similar for all three samples. A difference in the behavior of the mechanically activated micas samples arose during further heating, while there was no change in that of the initial sample. For the mechanically activated samples, it can be seen that the DTA curves slowly return toward the initial line, at a nominal revolution rate of the mill rotor of 20.000 r.p.m., sample (M_{12}), as a consequence of amorphization of the mica.

The X-ray analysis results of the mechanically activated mica samples of the series M_1, obtained in the mill with a circle sieve mesh of 80 μm and nominal rotor rate of 10.000 r.p.m., (M_{11}), respectively the samples from the series (M_{12}), at a nominal rotor rate 20.000 r.p.m., are shown in Figs. 5 and 6.

The X-ray analyses (Figs. 5 and 6.) indicate that changes in the structure of the samples appeared as a consequence of mechano-activation in the mechanical

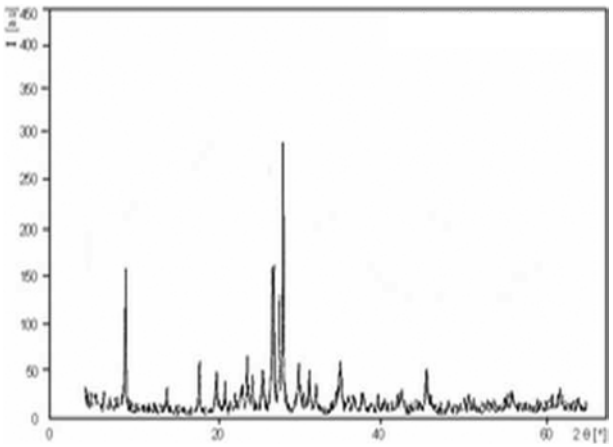

Fig. 5. Diffractogram of sample M_{11}

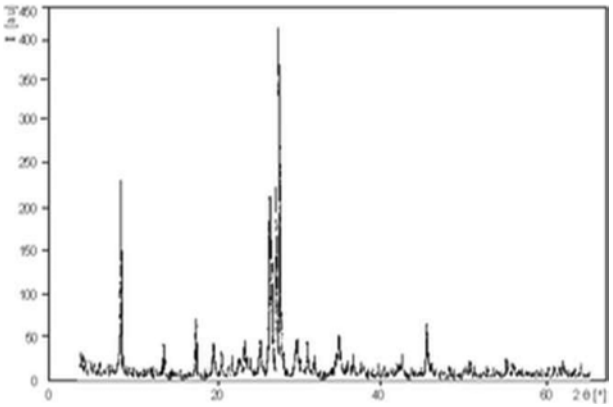

Fig. 6. Diffractogram of sample M_{12}

activation process. They are noticeable both qualitatively and quantitatively. The qualitative alterations of the phases present in the initial sample (mica, quartz, feldspar) functionally depend on the rate of rotor revolution and they are shown through the appearance of amorphous layers at the grains of the phases present. This amorphization process of the surface of the grains of the mechanically activated minerals depends also on their properties; hence for the mica (γ = 2.7 g/cm^3), it is very rapid in interval of n = 0-10.000 r.p.m., while for the hard minerals, quartz (γ = 7.0 g/cm^3) and feldspar (γ = 6.00 g/cm^3), the amorphization process is retarded.

This appearance can be explained by the mechanical activation of the mica grains and the release the quartz and feldspar inclusions in them, which naturally increased their concentration in the sample (M_{11}) at 10.000 r.p.m. In the case of the mechanically activated mica obtained at a nominal rate of the mill rotor of 20.000 r.p.m., sample (M_{12}), the effects of the process effects were slightly different. On the diffractogram of sample M_{12}, the intensities of characteristic reflections of quartz and feldspar were reduced, which indicates the commencement of their amorphization also. Unlike in the sample M_{11}, amorphous materials of quartz and feldspar composition were present in the sample M_{12}. From the quantitative aspect, the process is relatively simple, bearing in mind that the only process occurring is the transformation crystal phase amorphous phase.

Conclusions

According to the results obtained in this investigation of the kinetics of mechanical activation of mica in an ultra centrifugal mill, where the mechanical activation is realized firstly by impact under conditions of high mill rotor speeds, the best results were obtained at full mill load. The mechanical activation rate, as the main characteristic of mechanical activation kinetics, increased with increasing load and rate of revolution of the centrifugal mill rotor. The finest product in terms of massiveness was obtained with a circle sieve mesh of 80 μm and nominal rotor rate of 20.000 r.p.m. X-Ray and thermal analyses results showed that under these conditions of mechanical activation, the transformation process of a crystal phase into an amorphous phase occurred.

The results obtained in this investigation of mechanical activation represent a contribution to the interpretation of appearances and processes arising during mica mechanical activation in this type of mechanoactivator and may be useful for the introduction of these processes into practice.

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