Article

Assessment of Microsilica as a Raw Material for Obtaining Mullite–Silica Refractories

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Abstract: The possibility of using microsilica in the production of mullite–silica refractories was assessed. The chemical and mineralogical compositions of the raw materials, refractory Arkalyk clay and microsilica, were studied. It has been found that primary mullite and quartz formation occurs due to dehydration of kaolinite with the formation of intermediate metakaolinite. The introduction of alumina and microsilica into the charge composition promotes the formation of secondary mullite due to the interaction of aluminum oxide and highly dispersed chemically active microsilica. Free silica in compositions undergoes polymorphic transformations with the formation of cristobalite and tridymite. Mullite–silica refractories with an open porosity of 21%, a compressive strength of 42 MPa, and a thermal deformation temperature under the load of 0.2 MPa–1350 °C were obtained.

Keywords: aluminosilicates; mullite–silica refractory; microsilica; alumina; refractory clay; mullite; porosity; strength; structure

1. Introduction

The globalization of the modern world is only possible with the creation of various functional composite materials, including refractory and ceramic products.

The most common and popular type of refractory and ceramic products are aluminosilicate products, which include mullite–silica products containing 45–62% Al_2O_3. One of the most critical problems modern manufacturers of refractory and ceramic materials face is the creation of competitive products based on traditional raw materials and using secondary, artificial resources. The advantage of using technogenic raw materials is that there is no need for their extraction, which is expensive. Recycling them will reduce the cost of production and, at the same time, help improve the environmental situation in the region.

The main constituent phases of mullite–silica refractory and ceramic materials are mullite and silicon oxide. Natural and artificial alumina- and silica-containing materials are used to produce mullite–silica refractory products. There is an increased interest among researchers in using alumina- and silica-containing secondary and technogenic raw materials to create mullite functional materials [1–11].

For example, the authors of [6], in their work, used fly ash from thermal power plants to produce fused mullite powder with its further use as a filler for aluminosilicate refractory products. The introduction of orthophosphoric acid into the raw material mixture as a binder made it possible to reduce the firing temperature of fireclay refractory products by 150–200 °C, which made it possible to create an energy-saving technology.

Aluminum sludge was used in works [5–7,11]. It is a source of alumina used to produce mullite-forming refractory mixtures. The advantages of using aluminum sludge are the small particle sizes and less thermally stable properties of aluminum hydroxides, which

readily mix and react with silica to form mullite when sintered. When these compositions are heat treated, aluminum hydroxide is converted into γ-alumina and α-alumina.

Ceramic raw materials [5,6], glass waste [7], and slate waste [11] were added to the aluminum slurry as a silicon-containing component required for the synthesis of mullite.

There has been widespread use of microsilica as a silica-containing secondary raw material. Microsilica is a microscopic form of amorphous silicon dioxide. The crystalline form of SiO_2 is widespread and is found in almost all quartz rocks, but amorphous SiO_2 is practically never found in nature. Microsilica is a secondary product obtained in factory conditions. At the same time, the demand for microsilica in industry is steadily growing [12–17].

Microsilica is in most significant demand in the construction industry. Microsilica improves the quality of building materials produced [18–23] for reinforced concrete structures, various types of building mixtures, waterproofing materials, fire-resistant and fireproof materials, facing tiles, etc. For example, concrete structures can withstand heavy loads (compression of more than 80 MPa), anti-corrosion protection appears, frost resistance, and fire-resistant increase. The use of microsilica has also been noted in the production of thermal insulation materials [24,25]. A considerable amount of silica-containing products is formed during the acid processing of nepheline concentrate to produce alumina. The authors of [24] used these wastes to synthesize wollastonite with an acicular structure. Introducing wollastonite into ceramic masses reduced the temperature and duration of firing, reduced shrinkage, and increased the strength properties of materials. The authors of [25] used microsilica, which is a waste product from the production of crystalline silicon at a ferroalloy plant, to obtain high-modulus compositions. A granular thermal insulation material was obtained based on these compositions using a high-modulus liquid glass composition with a bulk density from 70 to 120 kg/m^3, compressive strength from 0.5 to 1 MPa, and water absorption of 7%.

The authors of [26] used exceptionally pure MS-971-grade microsilica produced by Elkem Materials to produce silicon carbide refractory products based on an alumina-containing binder. It has been established that the addition of microsilica in an amount of 2 wt.% (over 100%) ensures the preservation of a larger amount of silicon carbide in the refractory material and makes it possible to reduce the firing temperature of products from 1350 to 1300 °C and reduce energy costs to manufacture silicon carbide refractories. The use of microsilica ensures high quality, durability, and economic benefits for the materials that are produced.

Thus, an analysis of the available scientific information has shown that secondary raw materials and man-made waste, alumina- and silica-containing compositions, are widely used in producing aluminosilicate refractory and ceramic materials. It is noted that microsilica has recently attracted the most significant attention from researchers among silica-containing wastes. Microsilica is widely used in the construction industry. However, there needs to be more information on the research and use of microsilica to create refractory materials.

The sources of technogenic raw materials are mainly the mining, thermal power, and metallurgical industries. In Kazakhstan, during the production of metallurgical silicon at MK KazSilikon LLP, a huge amount of waste is generated during dust collection, which is converted into a by-product microsilica.

Purpose of the work: assessment of microsilica as a raw material for developing mullite–silica refractory.

2. Materials and Methods

The raw materials used in the work are refractory clay, technical alumina, and microsilica. The chemical composition of raw materials is specified in Table 1.

Table 1. Chemical composition of raw materials.

Raw Material	Content of Oxides, Mass. %								
	SiO_2	MgO	CaO	Al_2O_3	Fe_2O_3	K_2O	Na_2O	TiO_2	LOI
Refractory clay	37.5	0.1	0.82	39.7	1.85	0.3	0.09	2.7	16.94
Alumina	0.12	-	-	99.16	0.05	0.15	0.11	0.21	0.20
Microsilica	95.20	-	-	-	-	-	-	-	4.80

Refractory clay is from the Arkalyk deposit. Alumina is obtained by processing bauxite in Kazakhstan at the Pavlodar plant. Microsilica is a by-product of silicon production at MK KazSilikon LLP, Almaty, Kazakhstan.

The chemical and mineralogical composition of the starting materials and structural and phase transformations during heat treatment were studied with the use of X-ray fluorescence, X-ray diffraction, and thermal and microscopic analysis methods.

Refractory samples were obtained using methods adopted in the technology of refractory materials—grinding and fine grinding of starting materials, calculation and preparation of mixtures, preparation of molding mass, pressing from semi-dry masses on a hydraulic press, drying of samples, firing, determination of properties, structure, and phase composition according to the follow flowchart in Figure 1.
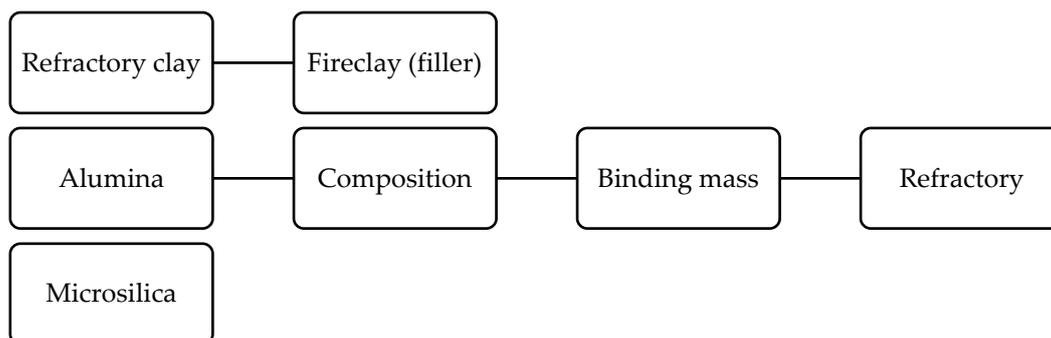
Figure 1. Process flowchart.

The properties of the samples were determined according to the following National standards: GOST 2409-80 "Fire-resistant materials and products. Method for determination of water absorption, apparent density, open and total porosity"; 4071-80 "Fire-resistant products. Method for determination of compressive strength"; 4070-2014 "Fire-resistant products. Method for determination of deformation temperature under load"; 390-2018 "Technical conditions. Fire-resistant fireclay and semi-acid products for general use".

Microscopic studies of samples were performed with MIN-8, Russia and OLYMPUS, Japan microscopes by transmitting light in an immersion environment and polished sections in reflected light using the Stream Program Basic R, Japan.

The phase composition of the compositions was studied using scanning electron microscopy (SEM) with electron probe microanalysis (EPMA) with an Optima 2000 DV atomic emission spectrometer, Perkin-Elmer, Waltham, MA, USA.

X-ray diffraction analysis of the starting materials and synthesized compositions was carried out with a D8 Advance diffractometer (BRUKER, Billerica, MA, USA), α-Cu radiation, and X-ray fluorescence on the Axios spectrometer, PANalytical, Almelo, The Netherlands.

Chemical analysis of the starting material was conducted in a specialized laboratory using certified methods with modern equipment.

3. Results and Discussion

3.1. Study of Raw Materials

The main mineral of the refractory clay from the Arkalyk deposit is kaolinite. The sample contained a small amount of gibbsite, quartz, and anatase (Figure 2).
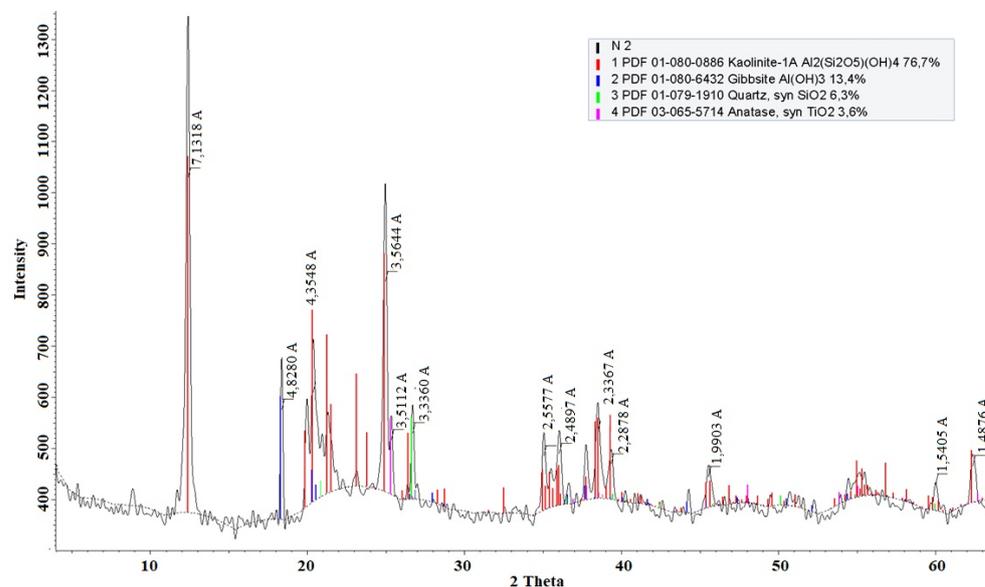

Figure 2. XRD pattern of refractory clay.

The DTA curve of the refractory clay sample shows endothermic effects at 119.2, 310.7, 560, and 575.2 °C. At the same time, they correspond to minima on the DTG curve at 95, 306, 559, and 566 °C. All of them are developed against the background of a decrease in sample weight. This means the gradual dehydration of kaolinite. The exothermic effect with a peak at 975.6 °C, developed against the background of a stable mass, can be interpreted as a manifestation of a restructuring of the intermediate metastable phase of metakaolinite (Figure 3). The endothermic effect with maximum development at 575.2 °C refers to the enantiotropic polymorphic transformation of quartz. The combination of endothermic effects with extremes at 310.7 °C and 575.2 °C reflects the presence of gibbsite. The fire resistance of Arkalyk clay was 1650–1700 °C.

Microsilica is a secondary product. It is formed during the high-temperature processing of silica-containing raw materials at MK KazSilikon LLP, associated with the sublimation process of silicon oxides. A finely dispersed colloidal, primarily amorphous material is formed when the sublimate condenses during the cooling process. The predominant particle size of microsilica is from 1 to 0.01 microns or less, which determines its high chemical activity (Figure 4).

According to X-ray phase analysis data, the microsilica sample is presented at (wt.%) 83.8 for the amorphous phase and 16.2 for the crystalline phase. The crystalline part is represented by mass.%: moissanite (SiC—64.5), cristobalite (SiO_2—20.2), and tridymite (15.3), which means that the total content in the sample is 10.4 moissanite, 3.3 cristobalite, and 2.5 tridymite. The amorphous phase of the product was represented by high-silica glass.

Based on the microsilica, samples were molded and fired at 1000–1500 °C in order to assess the behavior of this material during heat treatment. It was noted that with increasing firing temperature, amorphous silica microparticles sintered, forming aggregates. The silicon carbide and metallic silicon particles present remained unchanged. The structure of the samples fired at 1400 °C is porous (Figure 5a). In the structure of the fired microsilica samples, it was noted that the highly dispersed phase of amorphous silica was almost completely transformed into cristobalite and tridymite, and the spheroidal particles were sintered, forming aggregates.
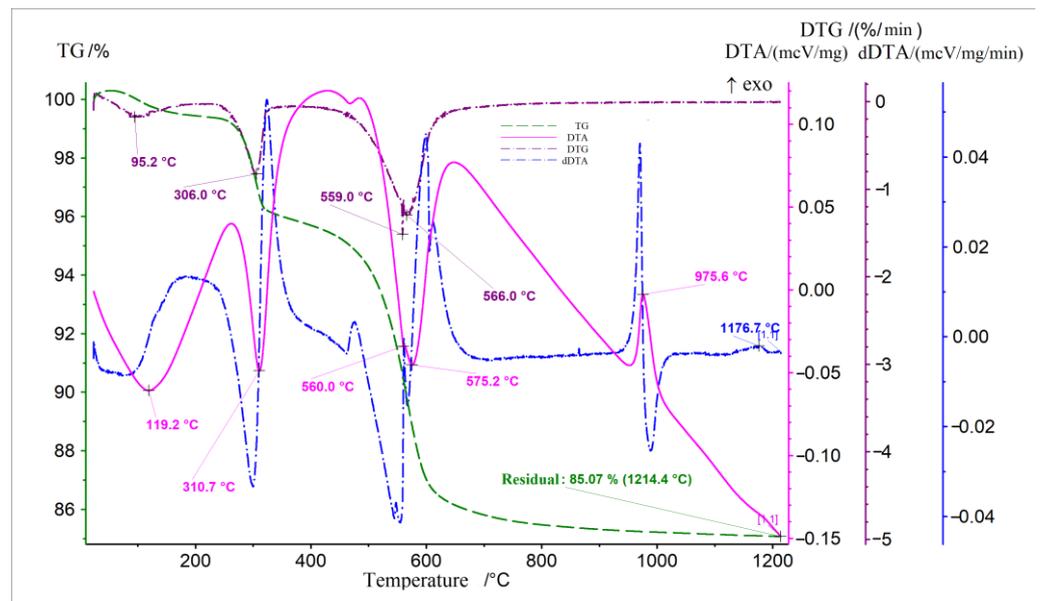
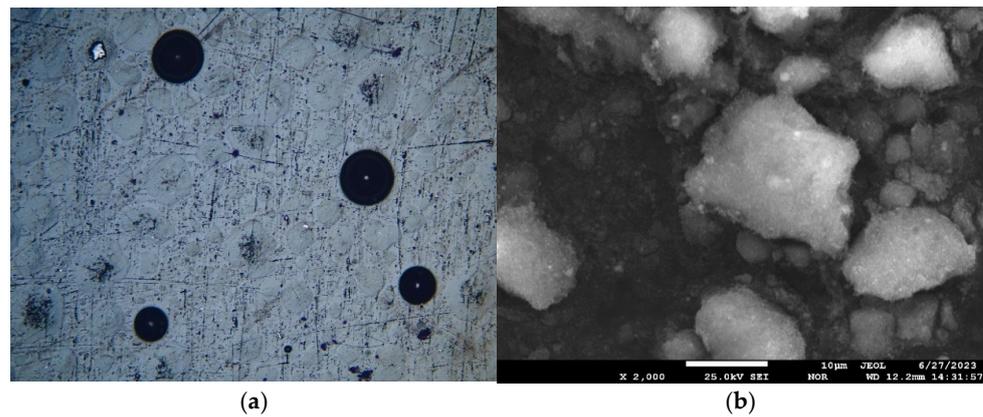**Figure 3.** Simultaneous TG/DTA analysis of the refractory clay.

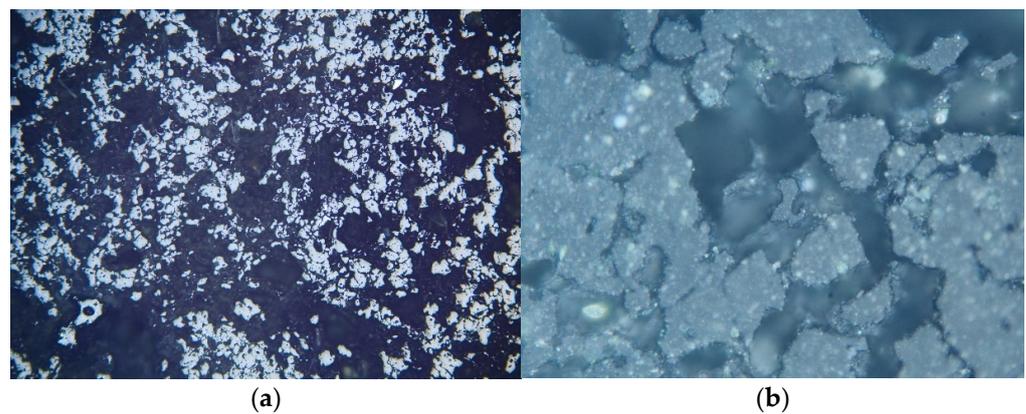**Figure 4.** Microsilica images: (**a**) reflected light ×50; (**b**) SEM image.

Figure 5. Structure of microsilica after heat treatment at 1400 °C. Reflected light. (**a**) ×50; (**b**) ×500.

The presence of inclusions of silicon carbide and metallic silicon (light inclusions) was also noted in the firing products (Figure 5b). The fire resistance of microsilica was 1600–1630 °C.

3.2. Selection of the Composition of the Refractory Binder

Traditionally, the mixture for producing refractory products consists of a filler and a binding matrix. The filler is a set of large fractions, and the binding part is made of a finely ground component containing a fraction of less than 0.063 mm.

The binding part of the refractories is most susceptible to aggressive action during the use of refractory materials in metallurgical units. Therefore, the best option is to create a refractory material where the phase composition of the binder consists predominantly of corrosion-resistant mullite.

In this regard, the work was devoted to selecting high-alumina compositions using microsilica for application as a binder for mullite–silica refractory products. The use of highly dispersed silica (microsilica) for the synthesis of a mullite matrix can be effective due to its developed surface and high chemical activity.

Refractory compositions have been formulated with varying ratios of refractory clay, alumina and microsilica. We calculated the chemical compositions (for a calcined substance) related to mullite–silica compositions to select compositions.

Calculations have shown that all compositions, except those based on refractory clay with the addition of 10 microsilica, meet the GOST requirements for Al_2O_3 content to produce mullite–silica products (Table 2).

Table 2. Chemical content of compositions.

Composition #	Composition of Mixture, Material, Mass.%	Content of Oxides, Mass.%							
		SiO$_2$	MgO	CaO	Al$_2$O$_3$	Fe$_2$O$_3$	K$_2$O	Na$_2$O	TiO$_2$
R10	refractory clay—100	45.14	1.20	0.98	46.80	2.23	0.31	0.10	3.24
R9	refractory clay—90 microsilica—10	51.45	0.11	0.87	42.31	1.96	0.32	0.10	2.88
R8	refractory clay—80 alumina—10 microsilica—10	46.12	0.12	0.76	48.37	1.74	0.28	0.08	2.53
R7	refractory clay—70 alumina—20 microsilica—10	40.98	0.13	0.65	54.20	1.53	0.24	0.07	2.20
R6	refractory clay—60 alumina—30 microsilica—10	37.14	0.72	0.59	57.84	1.36	0.24	0.10	2.01
R5	refractory clay—50 alumina—40 microsilica—10	32.64	0.60	0.50	63.08	1.15	0.22	0.10	1.71

Table 2 shows that compositions R5 and R6 have the highest aluminum oxide content and are more effective for obtaining mullite-containing compositions. Based on these compositions, samples were molded, followed by firing in the range of 1400–1600 °C to identify the patterns of structural and phase transformations that occur during heat treatment.

During heat treatment of compositions, structural and phase transformations occur associated with changes in the initial minerals and the formation of new phases based on them.

First, structural and phase changes occur in the clay component, which is the main one. At temperatures up to 1000 °C, dehydration of kaolinite is observed, with the restructuring of its structure into metakaolinite and the subsequent formation of primary mullite and quartz:

$$Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O \rightarrow Al_2O_3 \cdot SiO_2 + SiO_2 + 2H_2O\uparrow \rightarrow 3Al_2O_3\ 2SiO_2 + SiO_2.$$

The presence of these phases is established by the presence in the XRD pattern of lines of mullite (d = 3.38; 2.88; 2.69; 2.54; 2.20) and quartz (d = 4.26; 3.33; 1.83) (Figure 6). Studying

the samples of fired clay at a temperature of 1100–1200 °C with a microscope (in transmitted light) revealed the formation of a colorless, weakly anisotropic, uniaxial, negative phase in kaolinite with a refractive index of 1.55–1.58. The newly formed mineral with an imperfect structure is probably metakaolinite—a phase preceding mullite. A colorless, weakly anisotropic mineral was also noted in samples fired at 1400 °C, but with a higher refractive index equal to Ng—1.681, Np—1.661, biaxial, negative. This phase is the main one and, according to the X-ray phase analysis of the sample, is identified as mullite (d = 5.37; 3.38; 2.20) (Figure 6). In samples fired at 1400–1500 °C, recrystallization of mullite is observed, the crystals of which acquire a plate-like shape. The experimental results (Table 3) showed that samples based on refractory clay are sintered in the range of 1400–1500 °C. Increasing the firing temperature to 1600 °C leads to the appearance of some signs of burnout (deformation of samples, partial melting, decrease in density, and increase in open porosity). The optimal firing temperature for samples made from Arkalyk clay is in the range of 1400–1500 °C. A more detailed study of the structural-phase transformations that occur during heat treatment of Arkalyk clay is presented in works [27,28].
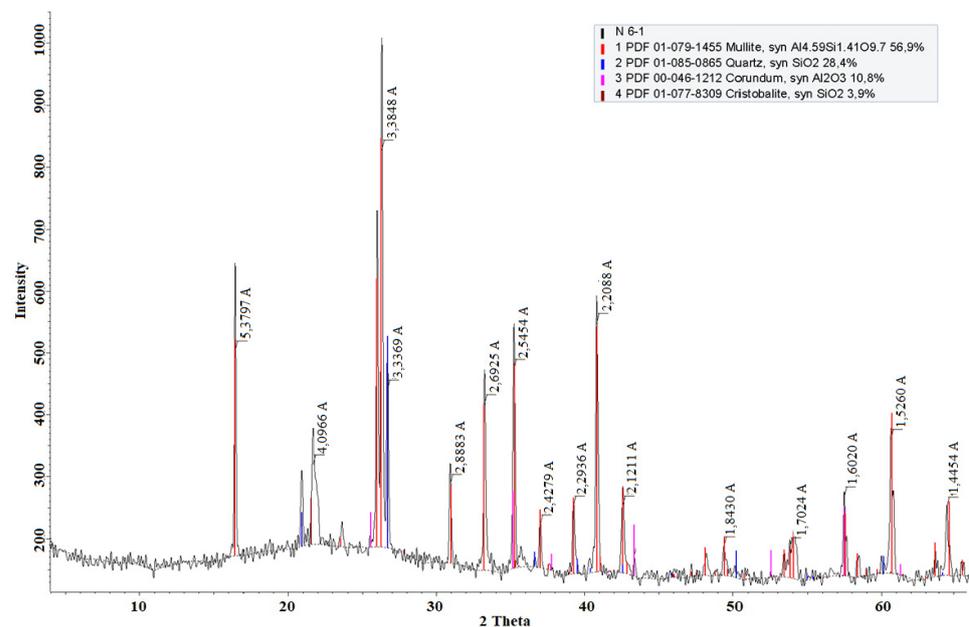

Figure 6. XRD pattern of fired refractory clay (1400 °C).

Table 3. Properties of samples of compositions.

Composition #	Firing t, °C	Open Porosity, %	Apparent Density, g/cm^3	Water Absorption, %	Compressive Strength, MPa
R10	1400	20.1	2.14	9.28	-
	1500	11.2	2.23	5.75	-
R9	1500	22.9	1.82	11.2	15.6
	1600	20.5	1.97	9.1	24.3
R6	1500	34.1	1.89	15.38	20.2
	1600	32.2	1.98	14.00	27.0
R5	1500	41.4	1.78	18.84	16.9
	1600	38.5	1.88	17.13	13.1

X-ray phase analysis of the fired clay (1400 °C) showed that the content of mullite is (approximately) 57, quartz is 26, and corundum is 10%, the rest being impurity minerals (Figure 6).

Introducing 10% microsilica into the composition can slightly change the phase formation process during the firing of samples.

Also, dehydration of the gibbsite occurs during heat-treating refractory clay. Gibbsite is also a source of aluminum oxide for the subsequent formation of mullite. First, primary mullite with an imperfect structure is formed from metakaolinite during the firing process. Aluminum oxide formed during the decomposition of gibbsite interacts with both silicon oxide of the starting material and microsilica introduced into the charge, forming secondary mullite. Due to the high activity of microsilica, the formation of secondary mullite with its participation is more likely. Moreover, microsilica interacts with raw material impurities, such as oxides of iron, titanium, magnesium, and others, with the formation of low-temperature compounds that form a eutectic melt of complex composition. The silicon carbide in microsilica does not change at the indicated temperatures.

The microstructure and phase composition of samples R9, R6, and R5 fired at 1500–1600 °C were studied using X-ray diffraction and microscopic analysis (Table 4, Figures 7–9).

Table 4. XRD results of compositions.

Composition	Phase	Formula	Quantity, %
R9	Mullite, syn	$Al(Al_{1.272}Si_{0.728}O_{4.864})$	53.1
	Cristobalite, syn	SiO_2	38.3
	Quartz, syn	SiO_2	8.5
R6	Mullite, syn	$Al_{4.68}Si_{1.32}O_{9.66}$	98.8
	Cristobalite, syn	SiO_2	1.2
R5	Mullite, syn	$Al_{4.68}Si_{1.32}O_{9.66}$	86.2
	Corundum, syn	Al_2O_3	13.0
	Cristobalite, syn	SiO_2	0.8
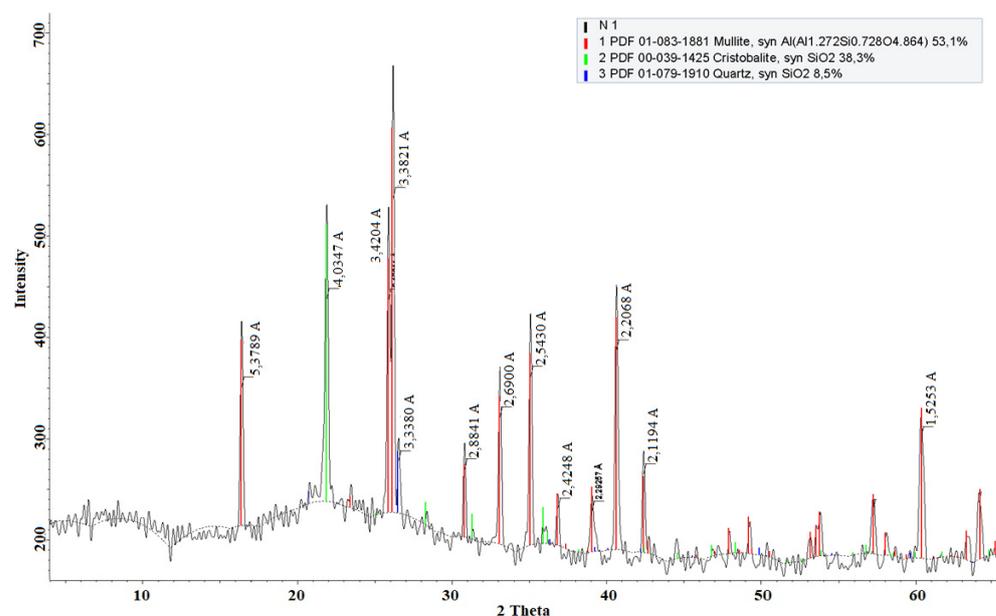
Figure 7. XRD pattern of the composition R9 (1500 °C).
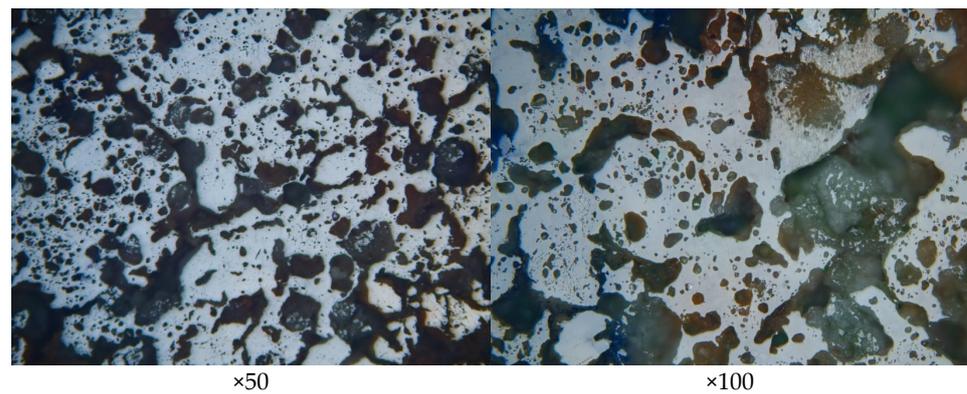
| ×50 | ×100 |

Figure 8. Microstructure of the composition R9 (1500 °C).
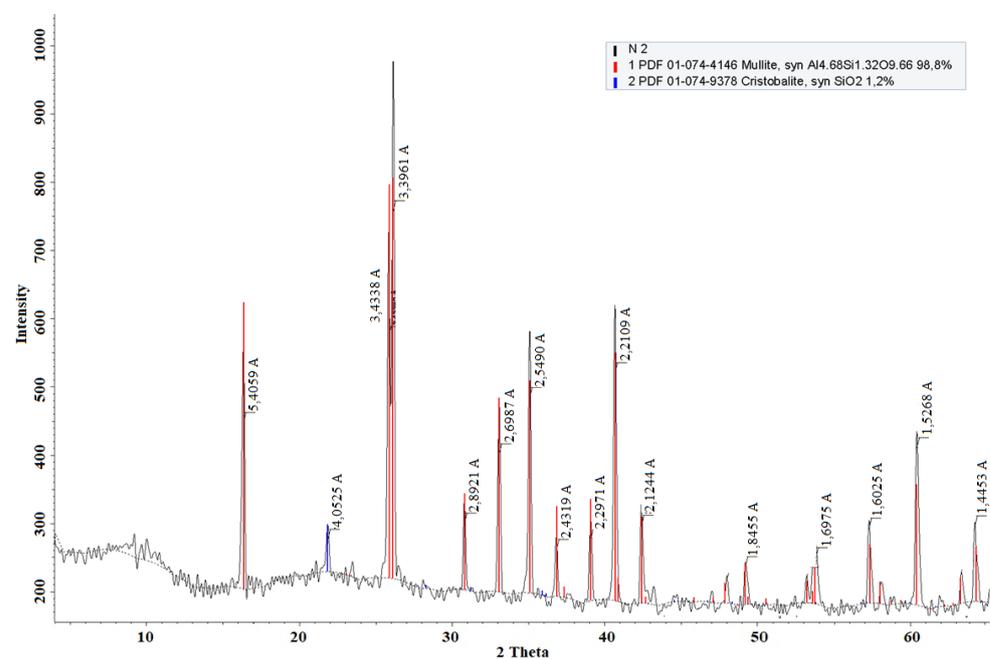

Figure 9. XRD pattern of a fired composition R6 (1600 °C).

Introducing alumina and 10% microsilica into the mixture increases the open porosity of the samples (Table 3, compositions R9 and R8).

The results showed that with an increase in the alumina content in the compositions, the values of open porosity and water absorption of refractory compositions increase, and loosening of the structure of the samples is observed. It indicates the intensive formation of mullite in the temperature range of 1500–1600 °C. Adding alumina to the charge increases the fire resistance of the compositions. For complete sintering and phase formation, increasing the firing temperature up to 1600 °C and increasing the soaking time are necessary.

The results of the X-ray phase analysis (Table 4) of fired samples showed that the mullite content increases with an increase in alumina additions. The most considerable amount of mullite was found in compositions containing 30–40% alumina and fired at 1500 °C. The mullite content in these compositions is about 86–98% according to X-ray phase analysis. However, the structure of the samples remains quite porous. It is required to increase the duration of the firing process at this temperature to compact the structure of the samples.

Increasing the firing temperature to 1600 °C affected the polymorphic transformation of quartz and microsilica into cristobalite, the content of which in the fired samples is approximately 1.2% (Table 4, Figure 9).

The structure of the sample R6 is porous and consists of mullite intergrowths. In the intervals, a silicate phase can be noted (Figure 10).
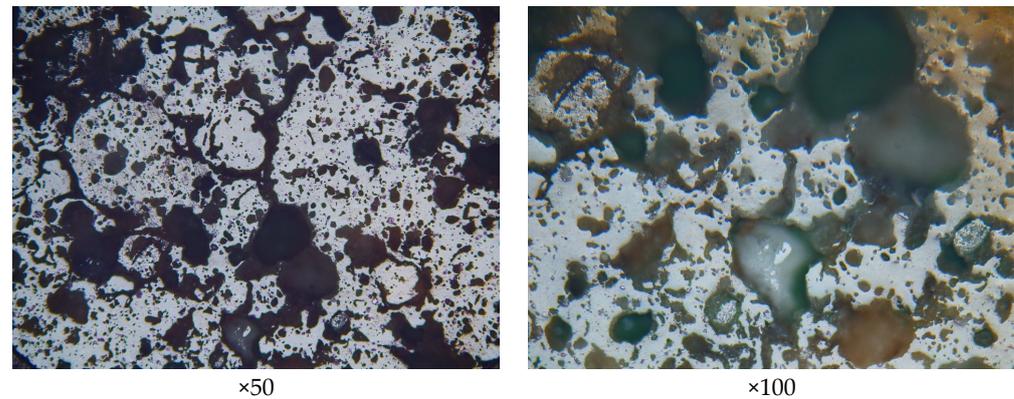

| ×50 | ×100 |

Figure 10. Microstructure of the composition R6 (1600 °C).

3.3. Obtaining Mullite–Silica Refractory

Mullite–silica refractory samples were obtained using traditional fireclay technology. The required filler fraction (less than 3 mm) was obtained as follows: refractory clay was briquette, dried, and fired at 1400 °C to produce fireclay. Preliminary firing of refractory clay into fireclay reduces shrinkage and improves the finished products' performance properties. The fired fireclay briquettes were crushed, ground, and fractionated into the required fractions (Table 5). The composition R6 and finely ground fireclay are used as a binding mass.

Table 5. Charge for obtaining mullite–silica refractories.

Compound	Material	Fraction, mm	Content, %
Filler	Fireclay	3–1	20
		1–0.5	30
		0.5–0.063	10
Binding mass	Fireclay	≤0.063	10
	R6	≤0.063	30

Lignosulfonate solution (1.23 g/cm^3) was used as a temporary binder. The molding humidity was 8–9%. Refractory samples were obtained at four specific pressures—40, 60, 80 and 100 MPa—on a hydraulic press to determine the optimal pressing pressure. The obtained samples were dried and then fired.

Molded refractory samples were dried at 100–105 °C and fired at 1500 °C. Fired refractory samples were tested to determine properties. The results are shown in Table 6.

Table 6. Properties of mullite–silica refractory samples.

Sample Pressing Pressure, MPa	Open Porosity, %	Apparent Density, g/cm^3	Water Absorption, %	Compressive Strength, MPa
40	22.73	2.12	9.70	32.6
60	21.29	2.16	8.98	42.1
80	24.12	2.08	10.38	36.7
100	25.17	2.05	10.94	32.3

Test results showed that the optimal pressing pressure is 60 MPa. The thermal deformation temperature under a load of 0.2 MPa was determined to be 1350 °C. X-ray phase

analysis showed that the main phase of the refractory is mullite, the approximate content of which is 86% (Figure 11). Impurities include corundum, quartz, and cristobalite. The structure of the sample is quite porous. Pores of various shapes are round, channel-shaped, and oblong (Figure 12).
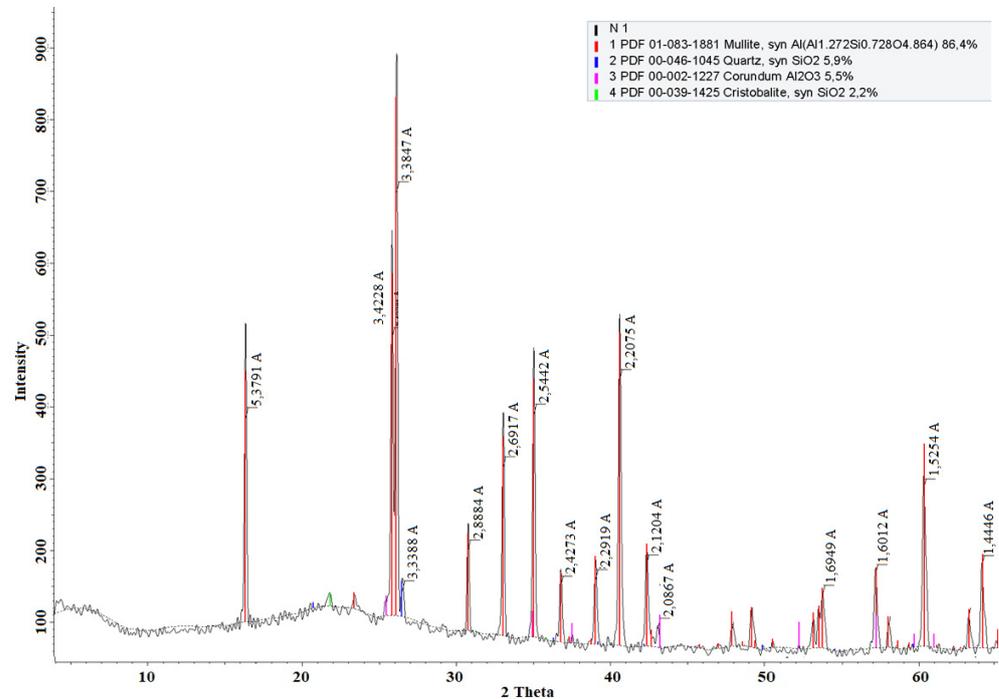

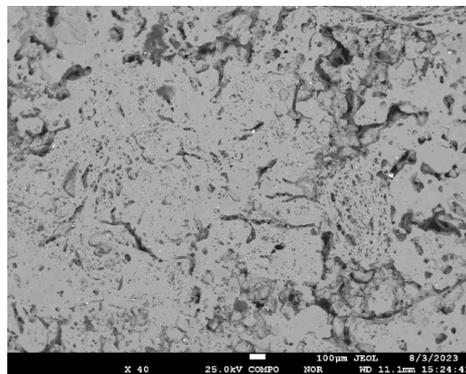**Figure 11.** XRD pattern of mullite–silica refractory (1500 °C, P = 60 MPa).

Figure 12. SEM image of mullite–silica refractory (×40).

The research results showed that the main phase of refractories is mullite, which determines the basic performance properties of products, such as strength, temperature at which deformation begins, corrosion, and thermal resistance.

4. Conclusions

Microsilica was assessed for its suitability as a raw material for producing mullite–silica refractory.

It has been established that the synthesis of mullite based on compositions containing refractory clay, alumina, and microsilica occurs primarily due to the decomposition of the main mineral of refractory clay—kaolinite. The introduction of alumina and microsilica into the composition promotes the formation of secondary mullite. Microsilica partially interacts with raw material impurities to form a glass phase of complex composition.

An optimal composition has been developed for producing mullite–silica refractory, consisting of fireclay filler and a binding mass consisting of refractory clay, alumina, and microsilica. It has been established that the optimal pressing pressure is 60 MPa. A refractory with an open porosity of 21%, a compressive strength of 42 MPa, and a temperature at which deformation began under load of 0.2 MPa–1350 °C was obtained.

Data Availability Statement: Some or all of the data or code supporting the results of this study are available from the corresponding authors upon request.

Conflicts of Interest: The authors declare no conflicts of interest.

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