Preparation of particle-reinforced mullite composite ceramic materials using kaolin and IG-017 bio-origin additives

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Abstract

In this research work, the possibilities of producing advanced ceramics from traditional, relatively inexpensive raw materials were studied. Powder mixtures from kaolinite and IG-017 bio-original additive were milled and uniaxially pressed to pellets, then pre-sintered in the electric kiln under oxidation and oxygen-free atmosphere at 1250 °C. The pre-sintered specimens were then further sintered in nitrogen gas at 1600 °C. The produced specimens were tested on composition, microstructures and morphologies using several characterization techniques including X-ray diffraction (XRD), scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDAX). The conducted experiments by the authors have shown that it is possible from traditional kaolinite to develop technical ceramic specimens with increased physical and mechanical properties based on formed amorphous phases, mullite crystals, micro and nano carbon fibers. Keywords: carbonization, ceramic, composition, microstructure, mullite, sintering

Kulcsszavak: karbonizáció, kerámia, összetétel, mikroszerkezet, mullit, szinterezés

1. Introduction

In recent years, there is a huge demand in many industries for new low-density materials with enhanced physical, electrical, magnetic, mechanical, thermal or chemical properties [1-34]. Thanks to these continuously increased requirements, more and more ceramic materials and ceramic-based composites are being developed and used in various fields of industry and technology [35-46]. Mullite is a stable intermediate phase under atmospheric pressure in the Al_2O_3 - SiO_2 system (Fig. 1) [47]. Despite the rare occurrence of mullite as mineral, the sintered mullite crystals are playing an important role both in traditional and modern technical ceramics. Fig. 1 shows how the various crystals and phases are depending on the temperature and also reveals that pure mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) melts at 1870 °C. Generally, at normal (atmospheric) pressure mullite crystals are capable to incorporate several transition metal cations and other foreign atoms into their structures depending on sintering environment and temperature [48-50].

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Fig. 1 $\text{SiO}_2\text{-Al}_2\text{O}_3$ phase diagram [47] (Taken from N M Bobkova: Fizicheskaya Himiya Tugoplavkih Nemetallicheskikh i Silicatykh Materialov, 2007)

1. ábra $\text{SiO}_2\text{-Al}_2\text{O}_3$ fázisdiagram [47] (Átvéve: N M Bobkova: Fizicheskaya Himiya Tugoplavkih Nemetallicheskikh i Silicatykh Materialov, 2007)

Generally, non-oxide technical ceramics and nitride ceramics have a much higher surface hardness and compression strength than metals and metal alloys or oxide ceramics, but their production costs are usually very expensive [51]. Nitride-based (SiAlON or Si_3N_4) materials are considered as significant and attractive structural ceramics/refractories because of their excellent properties, like high mechanical properties (e.g., strength, hardness, toughness), chemical inertness, good corrosion resistance and outstanding thermal resistance.

Silicon nitride is a non-natural mineral that needs to be synthesized by different methods [52-54]. The easiest way to produce silicon nitride from kaolin or other clay minerals is through carbothermal reduction and nitridation. The following reaction equations show the steps in which silicon carbide, silicon oxynitride, silicon nitride, and sialons are formed during the process [53-56].

1. Decomposition of kaolin at a temperature of 450 °C

2. Formation of mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) as compound of metakaolin at a temperature at 1087°C

While the temperature raises up to 1360°C, amorphous SiO_2 begins to transform into crystalline SiO_2 .

Carbothermal reduction and simultaneous nitridation occur at a temperature above 1400°C

3. Formation of silicon oxynitrides takes place progressively in environment richer in nitrogen under the reducing influence of the carbon

4. Formation of χ -phase sialon and β' -sialon from mullite

5. Formation of SiC

6. Formation of Si_3N_4

The Si-N bonds are formed at 1200°C in the presence of carbon. These bonds are progressively formed in N-rich environment and lead to a continuous formation of silicon oxynitride compositions. Usually, these bonds are not detected by XRD but appear later in the reaction, when crystalline silicon oxynitride persists throughout the reaction and possibly containing a little amount of Al (like O-sialon and x-phase sialon). After the destruction of the mullite, the Si-O bonds survive and remain throughout the reaction, especially in the kaolinite starting material. Later in the reaction, Si-C bonds are formed. The created SiC phase behaves more like a secondary product than a transitory intermediate [54-56].

Based on the studied literatures, it can be said that mullite or mullite containment taken from kaolinite can be successfully used as a matrix material for silicon nitride particles or whiskers reinforced ceramic composites. Si_3N_4 particles and whiskers can considerably enhance the mechanical strengths and toughness of both kaolinite and mullite based ceramic composites. Using conventional kaolinite or other types of clay minerals as starting materials, different micro and nano-size reinforcement materials can be produced like, SiC, Si_3N_4 , Si_2ON_2 or SiAlON through carbothermal reduction and nitridation.

The aims of this work are to develop ceramic composites material with superior physical and mechanical properties using relatively inexpensive processing technology and raw materials like conventional kaolinite or other clay minerals. In this work, the authors present a part of their research on the production of carbon-containing ceramics using kaolin and a biological additive. Further these produced ceramic composite could be a potential candidate for many applications showing better mechanical properties.

2. Materials and Experiments

Particle-reinforced mullite composite ceramics were prepared via carbothermal reduction and nitridation techniques, commercial kaolinite powders KKA-MB and IG-017 refined bio-original additives developed and produced by IGREX Ltd. were used as starting raw materials. Stoichiometric amount of these ceramic powders were weighted using 6 different mixtures compositions as shown in Table 1. The prepared ceramic mixtures were well milled in a laboratory ball mill for 20 minutes in the speed of 200 rpm. The milled powders then uniaxially compacted at 150 MPa pressures to produce 24 cylindrical disc specimens with diameters of 20 mm for each mixture. The filling weights of specimens were 8 grams in each case. After compacting, half of the specimens were pre-sintered in normal (oxidation) atmosphere and the other half of the specimens were pre-sintered in an oxygen-free atmosphere. In both cases, the maximum pre-sintering temperatures were 1250 °C. After these, the pre-sintered specimens were well studied and later they were burned at 1600 °C using nitrogen gas in the high-temperature kiln. Fig. 2 shows the sintered specimens.

Mixture	Kaolinite	IG-017
I.	85	15
II.	75	25
III.	65	35
IV.	55	45
V.	45	55
VI.	35	65

Table 1. The volumetric relationship between components in m%
2. táblázat Az EDAX vizsgálat során kapott elemi összetétel

Fig. 2 The specimens after the sintering process
2. ábra A szinterelt próbatestek

3. Results and discussions

Part of the authors have already shown in their earlier work some properties of the pre-sintered specimens, like volume shrinkage, weight losses, density, porosity and microstructure [57]. In this work are shown how the properties change due to sintering the specimens at a high sintering temperature (1600°C) in nitrogen gas. During pre-sintering of the specimens, firstly, drying of the samples takes place, followed by removal of the crystalline waters from kaolinite structure at approximately 450°C, parallelly gasification of the IG-017 additives occurs. Pre-sintering kaolinite at a temperature of 1250°C is high enough for the formation of mullite crystal phases and free SiO₂ components. Some of the specimens from each mixture were further sintered at 1600°C, where SiC and Si₃N₄ can be formed in the ceramic product. Figs. 3 and 4 show the volume shrinkages and weight losses of the sintered samples.

Fig. 3 compares the volume shrinkage of the heat-treated ceramic products to that of the pre-sintered and crude product volumes. Oxygen deficient pre-sintered ceramic specimens exhibit higher shrinkage. In contrast, the total volume shrinkages of the two type of pre-sintered specimens were almost the same.

Fig. 4 shows the weight loss of the specimens fired at 1600°C in an Oxygen atmosphere and Oxygen-free environment. In the case of the use of a larger amount of bio-original additive, the weight of the oxidation pre-sintered samples is increased, this could be possibly due to the incorporation of Nitrogen gas into the samples. In the case of oxygen-free pre-sintered specimens, some of the carbon remaining in the material structure is discharged during sintering at 1600°C. Examination of the microstructure and elemental composition of the samples was

done by scanning electron microscopy (SEM).

Fig. 3a
Fig. 3 The volume shrinkage after sintered the specimens at 1600 °C
3. ábra A térfogati zsugorodás az 1600 °C-os szinterelés után

Fig. 4a
Fig. 4 The weight losses after sintered the specimens at 1600 °C
4. ábra A tömegvesztés az 1600 °C-os szinterelés után

Fig. 5 shows the outer surface and breaking surface of a specimen made with 15 m% bio-additive pre-sintered in an oxidation atmosphere and then heat-treated in nitrogen at 1600°C. It can be clearly seen on the crushed surface that it has a high degree of porosity, from which the open pores and capillaries are significant on the surface. Based on the elemental composition determined by EDAX, the sample has a significant carbon and titanium at the surface, indicating that carbon and titan-containing surface layer has been formed during higher-temperature combustion. In the case of specimens with higher biological additive content, the surface of the pressed ceramic specimens was slightly nitrated.

Based on the EDAX test, the specimen contains 45 m% IG-017 has 1.51 m% nitrogen content (Table 2).

Fig 5a

Fig 5b

Fig. 5 The breaking surface (a) and outer surface (b) of the examined specimen from the I. mixture

5. ábra A I. keverékből származó minta belső, töretfelülete (a) és külső felülete (b)

The authors measured the oxide and phase composition of the sintered specimens with XRD test. After pre-sintering the specimens, the main phase was mullite with a large amount of amorphous glass phase and a small amount of quartz or cristobalite. During oxygen-free pre-sintering, carbon fibres were formed. After high-temperature heat treatment, the oxygen-free pre-sintered samples consisted of 60% mullite, 32% amorphous phase and 8% carbon fibres. Compared to the pre-sintered sample, the amount of carbon fibres in the sample was reduced (Table 3).

Fig 6a

Fig 6b

Fig. 6 The microstructure (a) and elemental composition (b) of the examined specimen from the IV. mixture

6. ábra A IV. keverékből származó minta mikroszerkezete (a) és elemi összetétele (b)

Mark of test specimen mixture		I.		IV.
Location of EDAX test		inside	surface	surface
Elemental compositions, in m%	N	0	0	1.51
	C	1.88	11.61	0
	O	24.00	32.81	31.81
	Na	0.47	0.54	0.81
	Mg	0.37	0.39	0.84
	Al	26.28	24.70	23.30
	Si	40.04	16.71	32.75
	K	4.59	1.15	4.33
	Ca	0.61	0.42	0.46
	Ti	0.67	10.98	2.83
	Fe	1.09	0.69	1.37

Table 2 The elemental composition based on EDAX test
2. táblázat Az EDAX vizsgálat során kapott elemi összetétel

Sintered atm.	Phase %	SUM	Mullite $\text{Al}_6\text{Si}_2\text{O}_{13}$	Quartz SiO_2	Cristo- balite SiO_2	Amorph glass SiO_2	Carbon fiber C
Oxidation		100	53	2	5	40	0
	SiO_2	61.95	14.95	2	5	40	
	Al_2O_3	38.05	38.05				
Oxygen-free		100	48	4	1	32	15
	SiO_2	50.54	13.54	4	1	32	
	Al_2O_3	34.46	34.46				
Nitrogen gas (1600°C)		100	60	0	0	32	8
	SiO_2	48.92	16.92			32	
	Al_2O_3	43.08	43.08				
	CO_2	29.31					29.31

Table 3 The oxide- and phase composition of the sintered specimens from IV mixture
3. táblázat A IV. keverékből készült minta oxid- és fázisösszetétele

4. Conclusion

In this research, the authors have successfully produced carbon-reinforced mullite ceramics using conventional kaolin minerals and IG-017 biological additive. Moreover, they continue their research on the production of mullite ceramics reinforced with silicon nitride particles or whiskers. Compared to the green products data, the weight loss and volume shrinkage of the prepared specimens after sintering at 1600 °C were not influenced by the pre-sintering atmosphere. However, the elemental and phase composition of the final products are significantly influenced by the type of applied pre-sintering process. In the case of specimens with higher biological additive content, the surface of the pressed ceramic specimens was slightly nitrated.

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