EXAMINATION OF THE CARBONIZATION PROCESS USING KAOLIN AND SAWDUST

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ABSTRACT

Ceramics reinforced with carbon and silicon carbide particles are of great importance in the field of technical ceramics and ceramic composites. In this research, the possibilities of producing advanced ceramics from traditional, relatively inexpensive materials were studied. The authors have successfully developed mullite ceramic materials reinforced with carbon particles using kaolinite powder and sawdust. Powder mixtures with different compositions were prepared in a planetary ball mill using silica balls. The produced mixtures were compacted in a 100 kN pull-press to produce pellets using a pressure of 140 MPa. The prepared specimens were sintered in a laboratory chamber kiln under oxygen deficient conditions at 1250°C. After sintering of the specimens, considerable amounts of carbon content were incorporated into the material structure of produced ceramics, which were confirmed by energy-dispersive X-ray analysis. Using 10–20 m% sawdust in the mixtures, the carbon (C) content of sintered specimens was found to be more than 8 m%.

Keywords: carbon, carbonization, ceramics, kaolinite, mullite, sawdust.

1 INTRODUCTION

Mullite and mullite-containing ceramics play an important role in both the traditional and technical ceramics industries. In the case of traditional ceramic products, the mullite phase is usually formed by the thermal decomposition of kaolin, while in the case of technical ceramics the mullite phase is formed by the reaction of the free quartz (SiO_2) and aluminium oxide (Al_2O_3) contained in the product. The phase purity and morphology of the mullite depend on the precursor materials and the methods of processing [1], [2]. Fig. 1 shows the SiO_2-Al_2O_3 binary phase diagram.

Pure mullite with high density cannot be produced by conventional sintering technologies. Usually it contains up to 10 m% of glass phase material and about 10% average porosity. Mullite ceramics that contain a small amount of glass phase usually have relatively high strength, low thermal expansion, so they have good heat shock resistance [1]–[6]. They have a better creep resistance than aluminium oxide ceramics. The properties of mullite ceramics can be advantageously influenced using metal oxides produced by mullite activation mechanism [7].

Based on the SiO_2-Al_2O_3 two-component phase diagram, the mullite is formed at 1087°C. Changing the ratio of oxides can highly influence the melting point of the product [3]. Starting from kaolinite minerals, tridymite is also formed from the metakaolin in the well-known thermal decomposition process along with the mullite [3], [8], [9], further increasing of the temperature results on conversion of tridymite into silicon carbide in the presence of carbon [10].

$$SiO_2 + 3C \rightarrow SiC(s) + 2\,CO(g). \tag{1}$$

Stoichiometric silicon carbide consists of one silicon and one carbon per unit compound. Silicon carbide has more than 100 known polytypes, the most important and best-known ones are hexagonal, rhombohedral and cubic crystal types as shown in Fig. 2 [11]. The SiC is often used as a reinforcing phase of various composites due to its high melting point, low density

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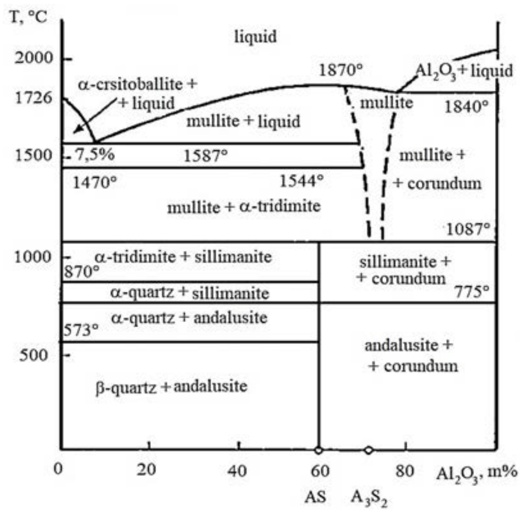
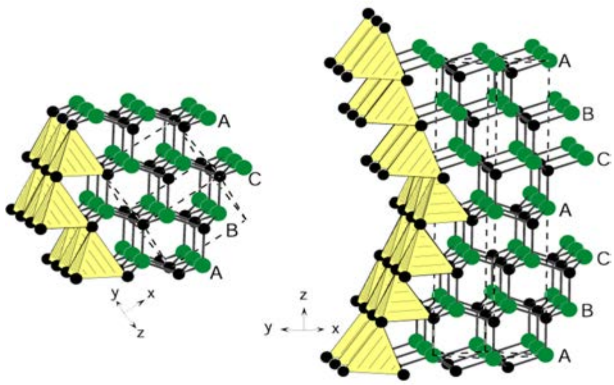


Figure 1: SiO_2-Al_2O_3 system at normal pressure [3].



(a) (b)

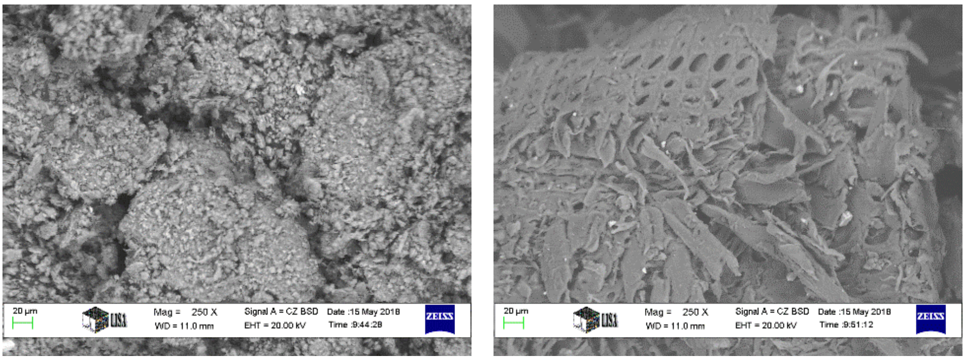
Figure 2: (a) 3C cubic β-SiC; and (b) 6H hexagonal α-SiC [18].

and high mechanical strength [12]. The SiC particles incorporated into the ceramic matrix properly improve the mechanical properties of the material [13]. SiC has excellent mechanical strength, good chemical resistance, high thermal conductivity, low thermal expansion coefficient and good heat shock resistance. It is therefore ideally used as a material for abrasives, filters, catalysts, acoustic and thermal insulation equipment, as well as a structural material or interior coating for high temperature furnaces [14]–[18].

The future aim of the authors is to develop ceramic composites with increased physical and mechanical properties using relatively inexpensive processing technologies and raw materials like conventional kaolinite or other clay minerals. The goals of this research are to prepare mullite reinforced with SiC or carbon nanoparticles and understand the influence of vegetable organic additive (sawdust) on properties of ceramic products including colors, microstructures, chemical compositions as well as shrinkages.

2 MATERIALS AND EXPERIMENTS

Zettlitz Sedlecky ml kaolin and fine grain oak sawdust (additive) were used as starting raw materials. Fig. 3 shows their microstructures. A suitable amount of kaolin and sawdust were prepared according to Table 1. The produced mixtures were milled in Retsch PM 400 planetary ball mill for 20 min at 150 rpm. The milled mixtures were uniaxially compacted at 140 MPa pressures to produce cylindrical disc specimens with diameters of 25 mm using 100 kN mechanical pull-press. Ten specimens of each mixture were made with a weight of 10 g.



(a) (b)

Figure 3: The microstructure of the raw material powders. (a) Kaolinite; and (b) Sawdust.

Table 1: Composition of the mixtures used in the research in weight percent.

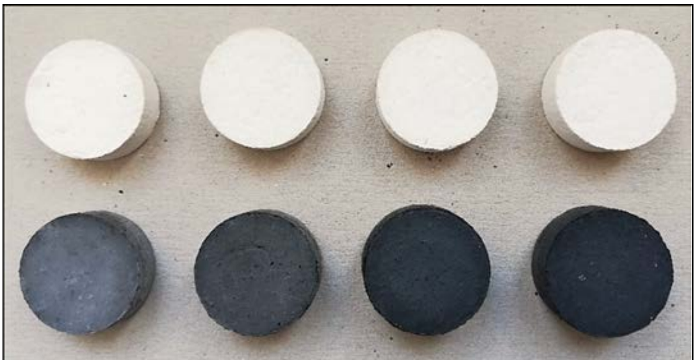
Mark of mixture	Quantity, m%	
	Kaolinite	Sawdust
A	100	0
B	95	5
C	90	10
D	85	15
E	80	20

The thermo-analytical properties of the powder mixtures were examined with a MOM Derivatograph-C. Raising the temperature, firstly leads to the drying of the sample, when all the moisture content of the mixtures leave. The next step is inflammation and burning of the additive (sawdust), which is most intensively carried out at 350–375°C, then, the thermal degradation of kaolin begins at 430°C. The kaolinite mineral ($Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$) produces metakaolin ($Al_2O_3 \cdot 2SiO_2$) when lose its crystal water [8], [9], [19]. This process is mostly occurred at 538°C. The process of mullite formation begins between 1000–1100°C.

After the thermo-analytical analysis and the compaction process the specimens were sintered in a laboratory chamber kiln at maximum temperature 1250°C and were kept at this temperature for 1 h. The sintering process of half of the specimens were made in a reduction environment in a sealed container.

3 RESULTS AND DISCUSSION

The geometrical parameters of the ceramic products are strongly depending on the material compositions and sintering conditions, like temperature, and sintering environment [5], [20], [21]. Due to sintering in different atmospheres, the colour of the ceramic specimens has changed as shown in Fig. 4.



5	10	15	20

Quantity of additive (sawdust), m%

Figure 4: The specimens sintered in oxidation atmosphere (white) and reduction atmosphere (black).

The effect of the applied heat treatment methods can also be observed by examining the volume shrinkage and the weight loss due to sintering (Fig. 5). Volume shrinkage of the pure sintered mullite specimens in oxidation atmosphere was found to be about 30 V% which slightly decrease with increasing the amount of sawdust. The weight loss of the sintered product has increased proportionally with the sawdust content of the green product. Volume shrinkage of the sintered samples in the reducing atmosphere is significantly reduced by increasing the amount of sawdust. However, the change of weight loss varies according to the curve with a lower slope than in the case of oxidation sintering. The reason is that, the additive (sawdust) has not completely burned out from the product. In case of 20% sawdust, the weight loss of the product in reducing sintering decreased by 5% less than that of oxidation sintering.

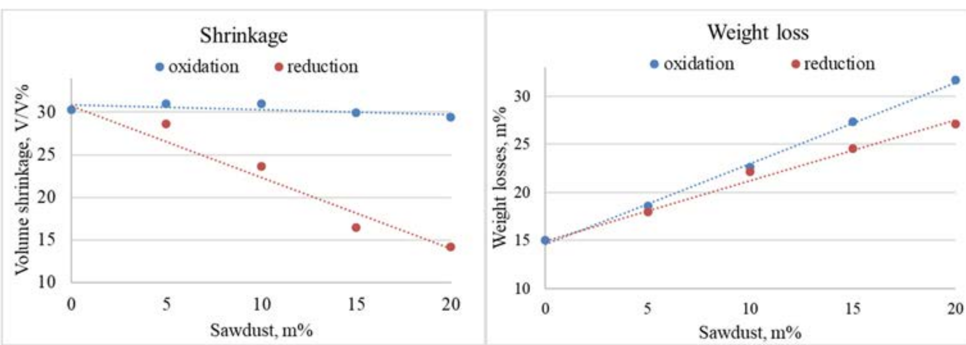


Figure 5: The volume shrinkage and the weight loss due to sintering.

The density of green-ceramic specimens was approximately 1.75 g/cm^3 which increased with oxidation sintering. Increasing sawdust content in all the cases reduced the density of the sintered product. The density of the produced ceramic samples can be seen in Fig. 6.

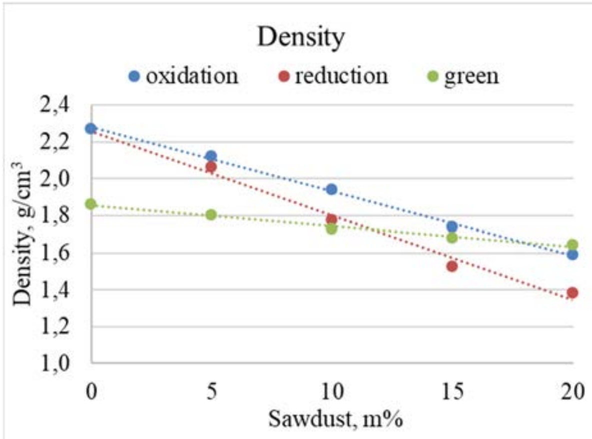


Figure 6: The densities of the produced mullite ceramics.

The microstructure of the powders and the sintered specimens were examined by Hitachi TM-1000 scanning electron microscopy. From the fracture surface investigation of the reduction sintered specimen made with 20 m%, it can be clearly seen that, sawdust is highly incorporated into ceramics structure (Fig. 7) [21].

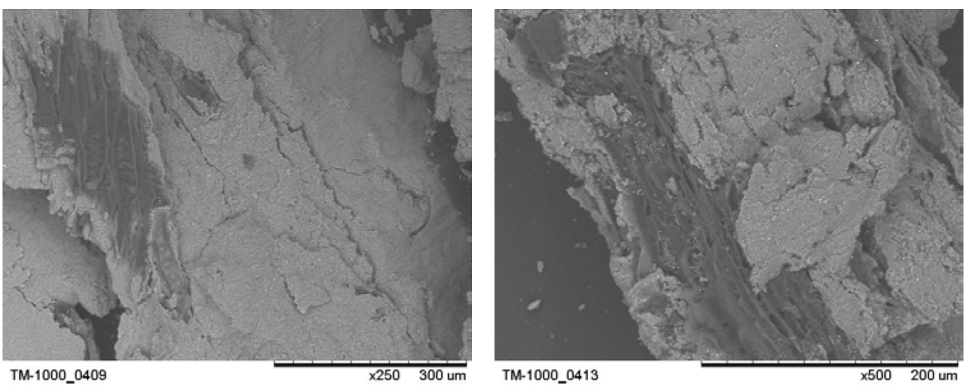


Figure 7: Fracture surface of the reduction sintered specimens with 20 m% sawdust.

The chemical compositions of the raw materials and the final ceramic products were examined by ZEISS EVO MA10 serviced with energy dispersive X-ray analysis (EDXA) micro-switch. Table 2 and Fig. 8 show the measured elemental compositions of the used raw materials and the fracture surface of the reduction sintered specimens. According to the data from EDXA, it can be observed that, increasing the amount of sawdust leads to increase the carbon content of the samples.

Table 2: Elemental composition of the used powders and fracture surface of the final products sintered in reduction atmosphere.

Element, m%	Powder		Fracture surface		
	Kaolinite	Sawdust	0 m% sawdust	10 m% sawdust	20 m% sawdust
C	–	62.15	–	8.58	14.89
O	45.36	37.25	38.80	34.56	28.85
Al	23.39	–	25.55	23.93	24.65
Si	29.14	–	32.18	28.80	27.81
K	0.83	–	1.04	1.05	1.38
Ca	0.47	0.60	0.49	0.61	0.47
Fe	0.81	–	1.94	2.46	1.95

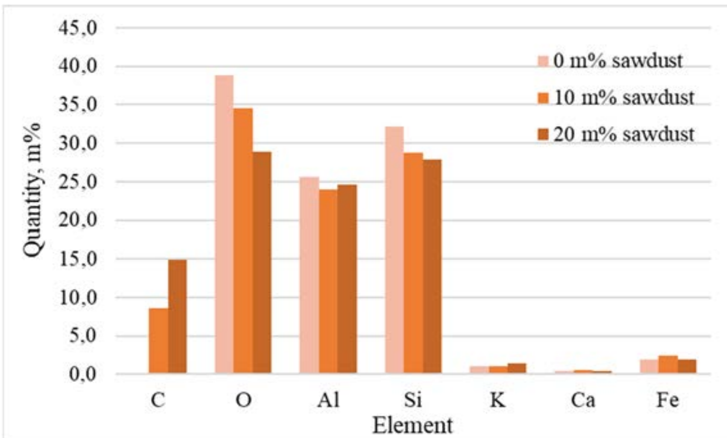


Figure 8: Elemental composition of the fracture surface of specimens of different mixtures sintered in reduction atmosphere.

4 CONCLUSIONS

From this research work we can conclude that, during the reduction sintering process a large amount of the vegetable organic additive (sawdust) was incorporated into the ceramic specimens. With increasing the amount of additive, large difference between the properties of ceramic specimens made by two sintering methods was noticed. The carbon (C) content of reduction sintered specimens was found to be more than 8 m% when 10–20 m% of sawdust was used in the mixtures. In this research work, the authors could successfully produce ceramic composites with increased properties using traditional kaolin and vegetable additives.

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