Development ceramic composites based on Al$_2$O$_3$, SiO$_2$ and IG-017 additive

E Kurovics$^1$, A Shmakova$^2$, B Kanev$^2$ and L A Gömze$^{1,3,4}$

$^1$Institute of Ceramics and Polymer Engineering, University of Miskolc, Hungary
$^2$Institute of Geology Komi SC UB RAS, Syktyvkar, Russia
$^3$IGREX Engineering Service Ltd, Igrici, Hungary
$^4$Tomsk State University, Tomsk, Russia

E-mail: mse205@gmail.com, femgomze@uni-miskolc.hu

Abstract. Based on high purity alumina and quartz powders and IG-017 bio-original additives the authors have developed new ceramic composite materials for different industrial purposes. The main goal was to fine a material and morphological structures of high performance ceramic composites as frames for development complex materials for extreme consumptions in the future. For this the mixed powders of Al$_2$O$_3$, SiO$_2$ and IG-017 bio-original additive were uniaxially pressed at different compaction pressures into disc shapes and were sintered in electric kiln under air (1) and nitrogen (2) atmosphere. The grain size distributions of the raw materials were determined by laser granulometry. There thermo-physical properties were also determined by derivatography.

The prepared and sintered specimens were tested on geometrical sizes, microstructure and morphology by scanning electron microscopy, porosity and water absorption. In this work the authors present the results of their research and investigation.

1. Introduction
In our days the industry requires more and more low density and mechanically strong materials capable to work in high temperature environment. To satisfy these inquires more and more ceramics [1-7] and ceramic matrix composites [8, 9] or cermets [10, 11]. To increase the wear resistance and live times of machine parts or machine tools the ceramic coatings and ceramic based composites are used in very often [12-14] Generally in cases when the machine parts and machine tools have to work under high mechanical loads and working temperatures the ceramic materials and ceramic composites are probably the best [15-18].

Our aims with this research work to create ceramic matrix composites and develop artificial meta-kaolin and mullite particles on the basis of different mixes of Al$_2$O$_3$, SiO$_2$ and IG-017 bio-original additives.

2. Materials and experiments
In our experiments we used alumina powders of Nabalox 315, silica flour from Fehérvárcsurgó and IG-017 refined bio-original additives developed and produced by Igrex Ltd. From these 3 components 18 different mixtures were prepared as it is shown in Table 1 and milled in laboratory planetary ball mill Retch PM 400 trough 20 minutes of each at 200 rotation/minute. During the grinding we did not take into consideration the mechnano-chemical phase transitions occurring under fine comminution. Later in a further work the determination of bond index of crushing can be also interesting using the easy method described by Maja S TRUMIC and her coauthors [19].

There were prepared by 6 different mixtures in the following 3 main combinations of components: I/ - Al$_2$O$_3$+IG-017, II/ - SiO$_2$+IG-017 and III/ - Al$_2$O$_3$+SiO$_2$+IG-017. From these 18 mixtures the
specimens were uniaxially compacted at 150 MPa pressures into shape of cylindrical discs with diameters of 20 mm. From each mixture we compacted by 10 specimens. The filling weights of specimens were 10 grams in each case. In spite of the same compacting pressures and filling weights we have got specimens of different heights (Figure 1) depending on material compositions of the used mixtures.

### Table 1. The volumetric relationship between the components

| Number of mixtures | I./ | II./ | III./ |
|--------------------|-----|------|------|
|                    | 1   | 2    | 3    | 4    | 5    | 6   | 1   | 2    | 3    | 4    | 5    | 6   |
| The composition in m% | Al₂O₃ | -    | -    | -    | -    | -   | -   | 90  | 80   | 70   | 60   | 50  | 40  |
|                     | -    | 90   | 80   | 70   | 60   | 50  | 40  | 31  | 29   | 27   | 25   | 24  | 22  |
|                     | -    | 80   | 70   | 60   | 50   | 40  | 31  | 29  | 27   | 25   | 24   | 22  |     |
|                     | -    | -    | 90   | 80   | 70   | 60   | 50  | 40  | 31   | 29   | 27   | 25  | 24  |
|                     | -    | -    | -    | 90   | 80   | 70   | 60   | 50  | 40   | 31   | 29   | 27  | 25  |
|                     | -    | -    | -    | -    | 90   | 80   | 70   | 60   | 50   | 40   | 31   | 29  | 27  |
|                     | -    | -    | -    | -    | -    | 90   | 80   | 70   | 60   | 50   | 40   | 31  | 29  |
|                     | -    | -    | -    | -    | -    | -    | 90   | 80   | 70   | 60   | 50   | 40  | 31  |
|                     | -    | -    | -    | -    | -    | -    | -    | 90   | 80   | 70   | 60   | 50  | 40  |
|                     | -    | -    | -    | -    | -    | -    | -    | -    | 90   | 80   | 70   | 60  | 50  |
|                     | -    | -    | -    | -    | -    | -    | -    | -    | -    | 90   | 80   | 70  | 60  |
|                     | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | 90   | 80  | 70  |
|                     | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | 90  | 80  |
|                     | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | -    | 90 |

After compacting half of the specimens were fired (pre-sintered) in normal (air) atmosphere and the other half of the specimens were fired (pre-sintered) in nitrogen (N₂) by the firing curves as it is shown in Figure 2. In both cases the maximum sintering temperature was 1250°C. This temperature is not enough high for formation of cristobalite crystals from tridymite phases in free SiO₂ components of the mixtures.

### Figure 1. Typical differences in heights of compacted items

### Figure 2. The sintering curves of specimens

3. Results and discussions

It is well known from practice that during sintering the Al₂O₃ – SiO₂ ceramics items are shrinking and this shrinkage very strong depends on firing conditions and temperature [20-23]. Our experiments confirmed that the geometrical parameters of the pre-sintered ceramic items strong depend not only on the material compositions but on sintering conditions, including temperatures, pressures and atmospherically environments, thanking to the chemical reactions and phase transformations in solid stages and gasification or degasification of volatile component (Figure 3).
The results well represent that most of the bio-origin IG-017 additives have fired out in normal (oxidation) atmosphere environment and the ceramic specimen suffered a large volume of shrinkage. This value of shrinkage is much higher than it could be expected because of the mullite formation from Al₂O₃ and SiO₂ components and phase transformation of the free SiO₂ particles from β-quartz to α-tridymite crystals. At the same time in nitrogen (N₂) atmosphere a swelling process can be observed thanking to the gasification and partly carbonization of the IG-017 additive micro and nano particles. Depending on volume ration of the used bio-origin additive during pre-sintering (green firing) some of specimens have shrunk (“+”) and some of specimens have swelled (“-“) both in normal and nitrogen atmospheres as they are shown in Figure 4. In cases when the specimens were made from all three powders (Al₂O₃, SiO₂ and IG-017) we could observe only shrinkage in the normal atmosphere (1 green line) and only swelling (2 green line) in N₂ atmosphere.

The burning weight losses (I) of specimens also were examined and determined by eq. 1.

\[ I = \frac{m_1 - m_2}{m_1} \cdot 100 \quad [\%] \]  

Where: the weight of the specimens in grams before firing (m₁) and after firing (m₂).

The burning weight losses of specimens are shown in Figure 5. The curves of this figure well illustrate that in cases when the portions of mixed IG-017 additives were less than 30 m% the burning weight losses of specimens were much higher in nitrogen (2) than in normal (1) atmosphere.

The water absorption (V_w) informs as about the open porous structure of the prepared ceramic items and can be determined as:

\[ V_w = \frac{m_\text{w}-m_2}{m_\text{w}} \cdot 100 \quad [\%] \]
Figure 5. Burning weight losses during green firing of specimens
1 – in normal (oxidation) atmosphere, 2 – in nitrogen (N₂) atmosphere

Where: the weight of the specimens in grams before \( m_s \) and after boiling \( m_w \) in water through 2 h.

The water absorptions of the green fired specimens as function of the volume of mixed IG-017 bio-origin additives are shown in Figure 6.

Figure 6. The water absorption of the green fired specimens
1 - fired in normal (air) atmosphere, 2 - fired in nitrogen (N₂) atmosphere

For the examination of the morphology and microstructure of the prepared specimens were used scanning electron microscopy and EDS. The typical microstructures and material compositions are shown in Figures 7-9 depending on quantity of the mixed IG-017 additives and the firing conditions.

Figure 7. Specimen prepared from III/5 mixture and sintered in normal (air) atmosphere
Figure 8. Specimen prepared from II/3 mixture and sintered in normal (air) atmosphere

Figure 9. Specimen prepared from II/3 mixture and sintered in nitrogen (N₂) atmosphere

The SEM pictures of II/3 specimens well illustrate that the ceramic items pre-sintered in nitrogen (N₂) atmosphere have much more and larger pores than the items fired in normal (oxidation) atmosphere. At the same time the items pre-sintered in nitrogen have much more small crystalline particles thanking to the reactive sintering and carbonization of part of the IG-017 additives and forming silicon carbide particles of micron and nano sizes. This formation of carbon base particles is well illustrated in EDS pictures, where the atomic percentage of carbon (C) has increased from 1.93 At% several times and achieved 8 At%. Thanking to the degradation of the IG-017 bio-origin additives, the volume of oxygen also has increased forming a new material structure of Si-O-C in solid phase.

4. Conclusions

Using normal (air) and nitrogen (N₂) atmosphere for pre-sintering were developed high porosity new ceramic specimens from alumina (Al₂O₃) powder, quartz flour (SiO₂) and IG-017 bio-original additives. The specimens were examined on shrinkage/swelling, water absorption and morphology. The achieved pore structures can make these ceramic materials suitable for high performance, light metal alloys impregnated new composites.

The formed new material structure of Si-O-C is much lighter than traditional Al₂O₃+SiO₂ ceramics and at high temperature it has an increased contact angel of wetness for most of light metal melts. Probably the developed by us new ceramic composite material composition and is pore structure can be impregnated with light metals and metal-alloys more easy and more perfect than the traditional ceramics and ceramic composites.

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