Development of a novel needle– like phase for alumina– doped glass with couple oxide addition

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Abstract

The present work aimed at investigating the use of (α-Al_2O_3) particulates in micro and nano scale; with the addition of soda-lime recycled glass powders to modify its sintering behaviors through following the role of the couple-oxide added like (Nb_2O_5-TiO_2) at different low concentration like 0.25 weight%, 0.5 wt%, 0.75 wt% and 1 wt%. The effect of alumina content on the ability to sinter at relatively low temperature was conducted for a series of Al_2O_3-glass composite samples having different concentrations of alumina, glass and additive couple oxides. The samples were prepared using ceramic technology route. Alumina weight was varied and takes the values 23wt %,33.5wt %, 44wt %, and 54.5wt % to which the oxide couple Nb_2O_5-TiO_2 was added and the samples were then denoted ASLG-NT. For all the prepared composite samples under study the sintering temperature was varied from 800°C to 900°C at a step of 25°Cwith a holding time of 2 hours at each temperature. The results showed that; as sintering temperature, and the couple oxides content increased together with the decrease in alumina content a relative density value of 98% was obtained. The Vickers micro-hardness measurements exhibited a value up to 42 GPa. Phase analysis conducted using XRD technique and microstructure analysis using SEM-technique revealed the development of a novel phase of needle-like grains starting from the surface towards the bulk which was of continues glass matrix in which the rest phase existed.

KEYWORDS: Bioceramic; Borosilicate glass; Biocompatibility

1. Introduction

Ceramic materials, mainly composed of metallic and nonmetallic elements like Al_2O_3, TiO_2 [1,2]. There are a lot of fields of applications of ceramics, glass ceramics and their composites in the biomedical domain because of their relevance with the physiological situations. Their hardness, strength and wear resistance of such Bioceramic like alumina and glass putting it at the front head of applications in dentistry where, ceramics are widely used for synthetic denture, abutments, bridges, dental fillings, crowns, implants and veneers over metallic foundations, because of their capability to imitative the optical properties of enamel and dentin as well as for their biocompatibility and chemical durability [3-7]. The concept of mixing two or more different ceramic oxides to obtain new materials with better characteristics has been applied for a long time. Glass - alumina composites, for example borosilicate glass with alumina, considered to be a good effort toward the use of such composites a base block for increased cost effective application due to their ability to density at low temperature and produce a desired properties [9,10]. In the present work; the role of couple oxide addition on sintering was adopted as a novel idea together with the use of recycled glass and nano-alumina powder was investigated in the present work.

2. Experimental

In this work soda lime glass used from recycled glass flat window glass which mechanically grinded using a special fine grinding machine (Retsch RS 200) then it was milled and sieved down to less than 30 µm, with α- aluminamicro particle of 50-200 µm from (Riedel- de Haen Germany) with added couple oxides (TiO_2-Nb_2O_5) which was prepared in the laboratory by wet mixing.The micro α- alumina by nano particle size with particle size of 30 nm and purity 99.5% (from S.S. Nano company, USA). Couple oxide, which was added at different weights (0.25, 0.5, 0.75, and 1) wt%and sintered at different temperatures 800, 825, 850, 875, and 900°C with 2 hours holding time. In order to attain good and desired homogeneity of the prepared samples, mixing was done in a planetary ball mill in which stainless steel balls of different sizes were used [large and small balls of diameters range between (0.5- 5) cm. Polyvinyl alcohol (PVA) alcohol was added to provide a wet medium for the milled powders to the grantee an even mixing of the prepared composite powders. The mixing was done in 24 hours at intervals of 6 hours each. The samples were then dried in vacuum oven at a temperature of 80 °C for 2hrs. A controlled amount of drops of polyvinyl alcohol (PVA) solution was added as a binder to the powders which gives a net of 5 wt% in the final mixture [11]. A pre-weighed samples of 1.30 gm each of the mixed powders were then pressed in a circular die to form pellets with 10 mm diameter and 5 mm height. Pressing was performed using a pressing pressure at (3.5) tons with a dwell time of 1minute, after which the samples were dried in an oven at a temperature (90) °C for 1hour. The samples were then placed in an electric furnace for sintering and the ramp rate used was 10 °C/ min.The samples were denotedaccording to weight as shown in table 1.

Table 1. Composition of alumina-soda lime glass composite with the couple oxides.

S.No	Materials	Wt (%)		Temperature (°C)
1-A 54.5 SLG-NT	Al_2O_3	54.5		800
				825
	Soda lime glass	45		850
	Nb_2O_5	0.25		875
	TiO_2	0.25		900
2-A 44 SLG-NT	Al_2O_3	44		800
				825
	Soda lime glass	55		850
	Nb_2O_5	0.5		875
	TiO_2	0.5		900
3-A 33.5 SLG-NT	Al_2O_3	33.5		800
				825
	Soda lime glass	65		850
	Nb_2O_5	0.75		875
	TiO_2	0.75		900
4-A 23 SLG-NT	Al_2O_3	23		800
				825
	Soda lime glass	75		850
	Nb_2O_5	1		875

	TiO_2	1		900

3. Results and discussion

3. 1 Effect of the couple- oxide addition on the densification of Nano/ Micro alumina

The highest relative density reached was for samples A 23 SLG-NTof nano and micro alumina soda lime glass to which Nb_2O_5-TiO_2 couple was added as 1wt %. For nano alumina a relative density of 98% and 97.2% for micro alumina at sintering temperature of 900°C with a holding time of 2 hours. As the sintering temperature increased from 800 to 875 °C the density was increased with increasing the temperature for all the various specimens that were shown in figs.(1) And (2).The degree of densification was affected by the amount of the couple – oxide Nb_2O_5-TiO_2 added, a small amount of dopants does not seem to improve the degree of densification of alumina at lower sintering temperature, but the relative density of doped samples significantly appeared to increase with increasing sintering temperature and the amount of the couple oxide. The measured density of all samples shows that there was no large difference between the two types of nano and micron Al_2O_3 particle size in the relative density. The sintering additives were noticed to have an obvious role to improve the ceramic material density and get a controlled grain structure and desired phases.

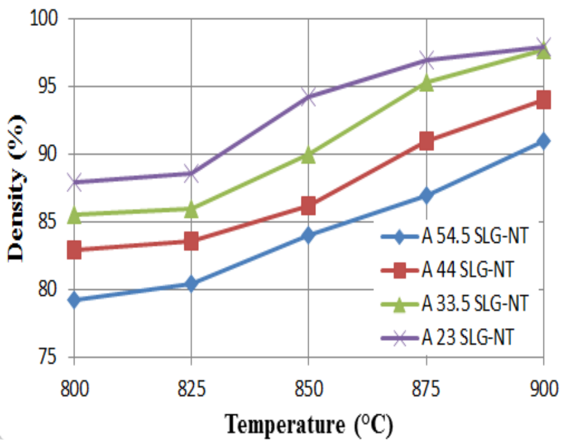


Fig. 1; nano α- AL_2O_3–soda lime glass with Nb_2O_5-Tio_2 additiveeffect on the relative density.

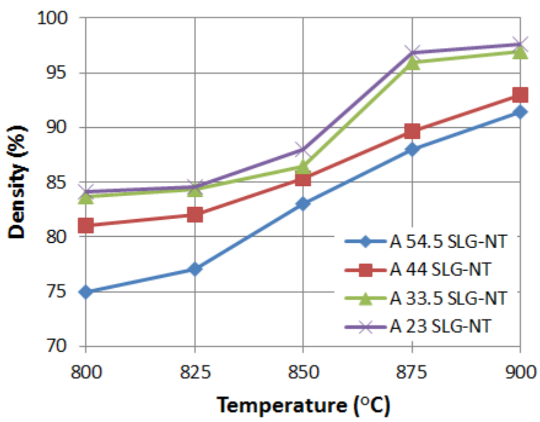


Fig. 2; micro α- AL_2O_3 –soda lime glass with Nb_2O_5-Tio_2 additive effect on the relative density.

3. 2. Effect of Sodalime glass - oxide couple additives on the hardness of Alumina composites.

The Vicker hardness of micro alumina in the sample seriesA 23 SLG-NT was shown in the fig (3) and a maximum value of 25.4 Gpa was exhibited at 900°C with the couple additives of 1wt%, but for sample with 44 wt% of α-alumina the hardness was less than the first composition which has the value of 21.8 GPa. According to these results such behaviors could be referred to describe at least three factors affected on the hardness; like additive oxide content, temperature, the amount of alumina, and also the density of the samples. (Fig. 4) showed two different amounts of Al_2O_3 which was 44 and 23wt % of Nano alumina that were added to soda lime glass with the couple oxide Nb_2O_5-TiO_2

additive. The sample at 23wt% shows, the highest degree of hardness among all the samples which was 42 GPa, and for 44wt% of nano alumina hardness value was 40 Gpa. This value of hardness was closely comparable with that of the tooth and match with those by P. Gutiérrez-salazar and J. Reyes-gasga [12]. Such results give an indication that as the alumina content decreases the hardness value increased which conforms with the important role played by the temperaturewhich highly effect on the sample properties. In addition to that the SEM micrographs showed the appearance of needle- like shapephase precipitated, which was observed dispersed within the matrix. This phase considered as a new feature of the prepared microstructure which was noticed for the other samples with Nano- alumina powder.

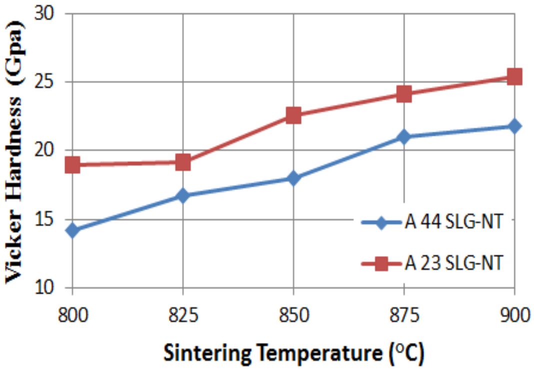


Fig. 3; Effect of addition soda lim glass with additive couple oxide (Nb_2O_5-TiO_2) to micro size α- Al_2O_3 on the vicker

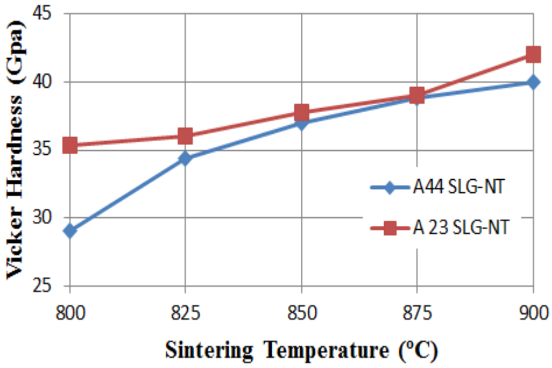


Fig. 4; Effect of addition soda lime glass with additive couple oxide (Nb_2O_5-TiO_2) to Nano size α- Al_2O_3 on

3.3. XRD analysis for nano/ micro alumina with soda lime glass and couple oxides (Nb_2O_5-TiO_2)

The X-ray diffraction for using nano alumina in A 23 SLG-NT sample as shown in Fig. 5 with couple oxides additionsof 1 wt% The phase analysis by XRD reveals the formation of α-Al_2O_3 as in card (46-1212), SiO_2 card (46-1242), and $Na_2Ca_4Mg_2Si_4O_{15}$ according the card number (42-1484) Moreover the appearance of a novel new phase called Devitrite ($Na_2Ca_3Si_6O_{16}$) which developed (23-0671) with increasing sintering temperatures was checked also by SEM in addition to XRD.The analysis reveals the formation of a needle like morphology, which expected to make this sample taking the highest hardness and density values.Also phase analysis of 23wt% for micro alumina was showed the appearance of Al_2TiO_5although limited, but at high intensities as in the card number (09-0252). Moreover, other phases like $Na_2Ca_4Mg_2Si_4O_{15}$ card number (42-1484), Al_2O_3 card (42-1468), and trace of Nb_2O_5 phase were also present as shown in (Fig. 6).

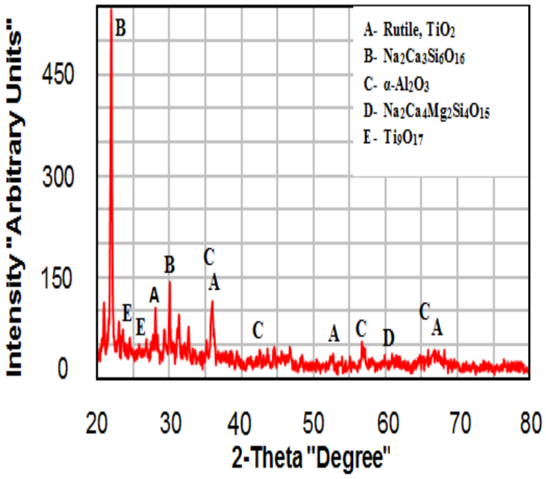


Fig. 5; XRD analysis for micro alumina 23wt%with soda lime glass and 1wt% Nb_2O_5-TiO_2

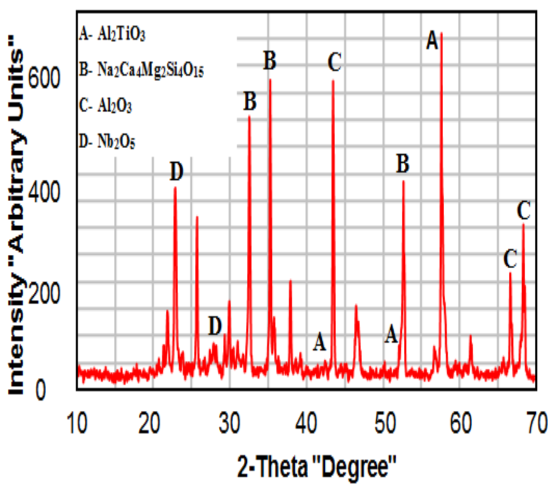
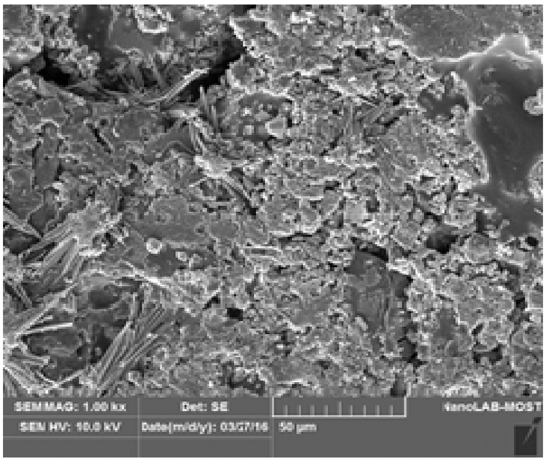
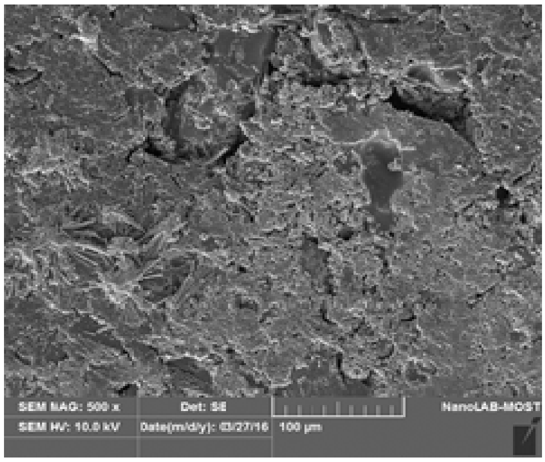


Fig. 6; XRD analysis for nano alumina 23 wt% soda lime glass with 1wt% Nb_2O_5-TiO_2

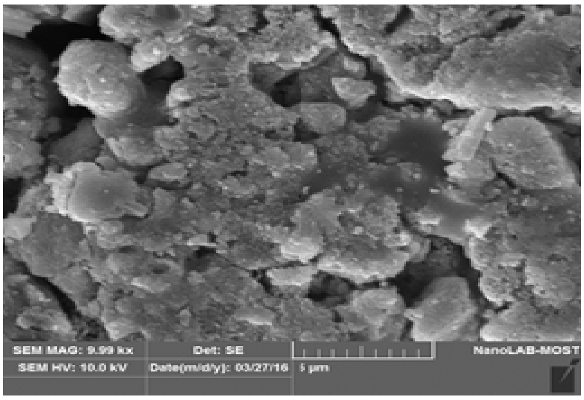
3.4 .Microstructure Analysis by Scanning Electron Microscope (SEM)

(Fig. 7) Shows SEM micrographs using Nano-Alumina for samples sintered at 900^{o}C. From these micrographs a high percent of fused areas (glassy phases) that is the main phase for high sintering temperature which will cause an increase in the density of the sample. Deep analysis of the SEM micrographs in (Fig. 5-26- A, B) showedthat certain parts of the glass bond had been de-vitrified and the crystals were grown and takes some shape like elongated section. In addition, SEManalysis showed the presence of one direction growth of dispersed areas in the samples resulting in a needle- like and to lower extension a rod- like morphology of the resulted phases. This behavior could be ascribed to the direct effect of the added oxide couple in the presence of Nano alumina which catalyzed the crystallization of definite phase like devitrite (sodium- calcium- silicate) together with the growth of Nano alumina particle to take the rod- like morphology. A second crystalline phase composed of titania- doped aluminum oxide structures was also noticed in the microstructure of the sintered samples. The SEM and XRD analysis also revealed the microstructure of the tested phase consists of needles of rutile and rutile- like (Ti_9O_{17}) orientated on the faces of titanium-doped aluminum oxide grains that penetrate into the glass bond[13-15]. With the increase in TiO_2 content, more plate formation is visible in the structure, that may explain needles formation and caused by the increased of TiO_2 percent of this sample [16]. When the micro alumina powder used to prepare samples processed under similer preparation conditions the needle- like morphology was disappeared as show in (Fig. 8). Due to use of micro alumina and the sintering temperature used does not enough to create a condition suitable for the formation of the needle shaped phases As shown in (Fig. 8- A, B) because there was no enough activation energies for sintering and growth of such phase, which was indicated by the low densification noticed for those systems. Microstructure investigation reveals the presence of coarse and fine grains with more porosity as appeared in the SEM microgrph at (Fig. 8- c).



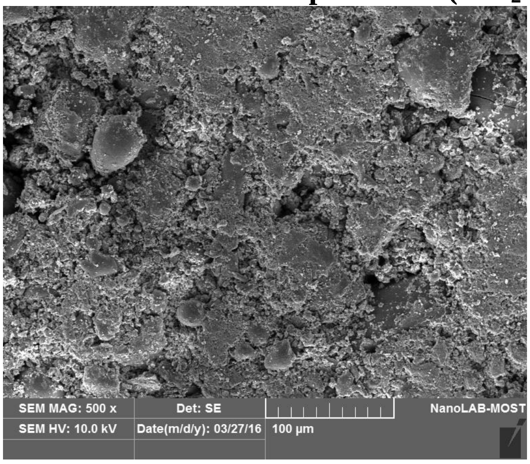


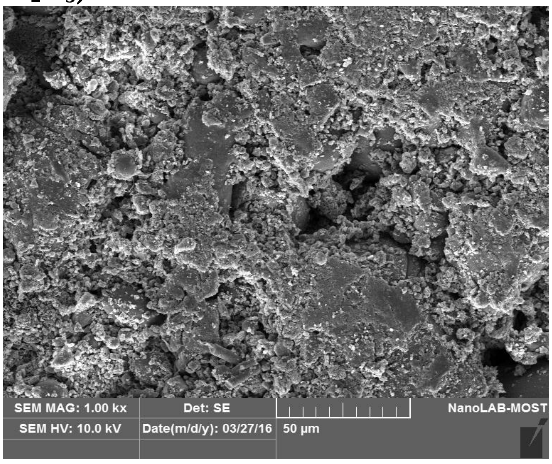
A B



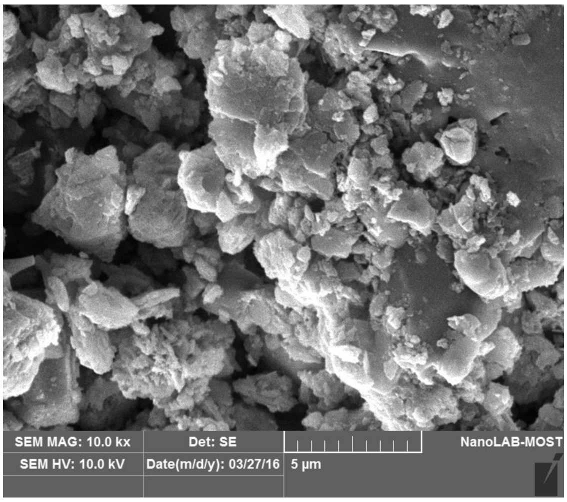
C

Fig. (7-A, B, C); SEM micrograph for nano α- alumina 23 wt% soda lime glass with with 1wt% for couple oxides (TiO_2- Nb_2O_5)





A **B**



C

Fig. (8-A, B, C); SEM micrograph for micro α- alumina 23 wt% soda lime glass with with 1wt% for couple oxides (TiO_2- Nb_2O_5)

4.Conclusions

1- The intrinsic properties of alumina exert a major effect on the whole properties of the prepared composite. As it is content increased; densification and mechanical properties were relatively hindered by the formation of intergranular porosity and / or secondary phase.

2- Addition of nano alumina makes the densification rate more linear for all compositions under study as compared with micro-sized alumina when Al_2O_3 content decreased from 55-23 wt%. Using the Nb_2O_5 – TiO_2 additives couple. Moreover the densification curves for micro alumina composite were noticed to converge closely which was not the case with micro-Al_2O_3.

3- Addition of alumina in general and specifically nano-alumina raised the refractoriness, density and hardness of the composite bodies.

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