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Effect of Addition of B_2O_3 on Dielectric Properties of BMN, BZN, BMNT and BZNT

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ABSTRACT: Niobium and tantalum-based ternary oxides have been found to be good candidates for electronic applications because of their excellent dielectric properties.In earlier studies it was found that addition of frit glass improves density, mechanical strength and dielectric properties of BMT hence, in the present study B_2O_3 was added to BMN, BZN, BMNT andBZNT. These samples were prepared by solid state route and the phase was confirmed by XRD. To these samples B_2O_3 was addedin varying weight percent (1 to 3%) and the pellets were sintered at 1250˚C. These samples were subjected to capacitance and dielectric measurements. From the results it was observed that addition of frit glassincreased the density to some extent but did not improve capacitance and infact reduced the dielectric constant drastically of the ceramics. Hence it is concluded that addition of B_2O_3 has detrimental effect on BMN, BZN, BMNT and BZNT.

KEYWORDS: Niobates and Tantalates, B_2O_3 additive, dielectric properties.

I. INTRODUCTION

Microwave dielectric ceramics with high dielectric constant ε_r(20-30), high quality factor Q (1, 00,000 to 1, 50,000) and near zero temperature coefficient τ_f are used as key materials in communication systems [1].Ternary niobates and tantalates of various metals are widely used in contemporary technology. In wireless communication system at microwave region, such materials are of immense interest due to their applications as filters, oscillators, dielectric resonators in mobile phones communication, and satellite communicationstechnology[2]. Several niobium and tantalum based dielectric oxides with perovskite structure have been found to have potential applications in communication technology [3-5]. However, search for new compounds with optimal dielectric properties continues to be of great interest. As a logical continuation of our work on BMT and BZT,we started the present work on BMNT and BZNT with a hope that dielectric constant can be kept in the range of 20-30 and still cost reduction can be achieved by substituting tantalum with niobium.

In earlier studies, it was found that BMT with 2.5% of frit glass as additive gave highest theoretical density to the ceramic [6]. The present study was carried out on this basis so as to achieve highest theoretical density to BMN, BZN, BMNT and BZNT.

II. EXPERIMENTAL

BMN, BZN, BMNT and BZNTwere prepared using a conventional solid-state reaction method. Powders of $BaCO_3$, MgO, ZnO, $Nb_2O_5$and Ta_2O_5 were used as the starting materials. Starting materials were weighed according to stoichiometric ratio (3:1/1:1) so as to give final compositionas$Ba_3(MgNb_2)O_9$, $Ba_3(ZnNb_2)O_9$, $Ba_3(MgTa_{1.8}Nb_{0.2})O_9$ and $Ba_3(Zn\ Ta_{1.8}\ Nb_{0.2})\ O_9$.The raw mixtures were then ball milled for 2 h with iso-propanol using zirconia balls as grinding medium. The mixtures were calcined at 1300°C for 2 hours.These samples were then pressed to form pellets of 10mm diameter and about 4 to 5 mm thickness using 5% poly vinyl alcohol as binding medium at a pressure of 5 tons for two minutes. The pellets were heated slowly up to 600°C to remove the binder and were sintered at 1250°C for 2 hours. In the similar manner pellets of the above mentioned calcined ceramic samples with 1% and 3% frit glass (B_2O_3) by weight were prepared and were sintered at 1250°C for 2 hours. The powder samples were tested by X-ray diffractometer (Model: Bruker

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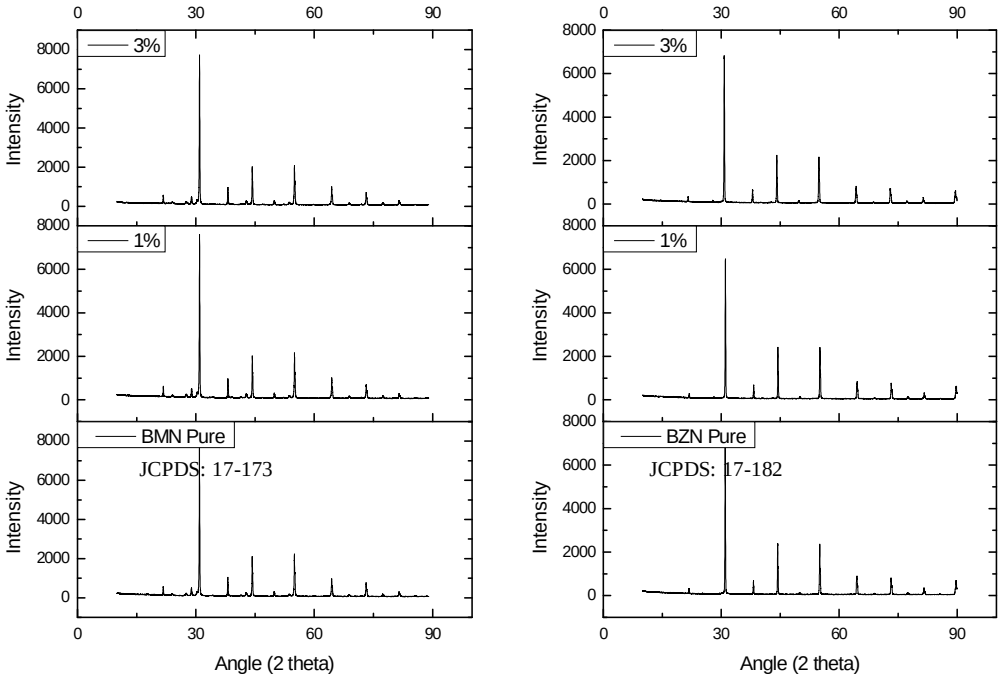
(An ISO 3297: 2007 Certified Organization)

Vol. 3, Issue 6, June 2014

D8 Advance) for the crystal structure and the sintered pellets were tested by SEM (Model: JEOL JSM 6360A) for surface morphology.The capacitance was calculated on 4284A Precision LCR meter.

The XRD patterns of the ceramics with and without frit glass are shown below:



(a) **(b)**

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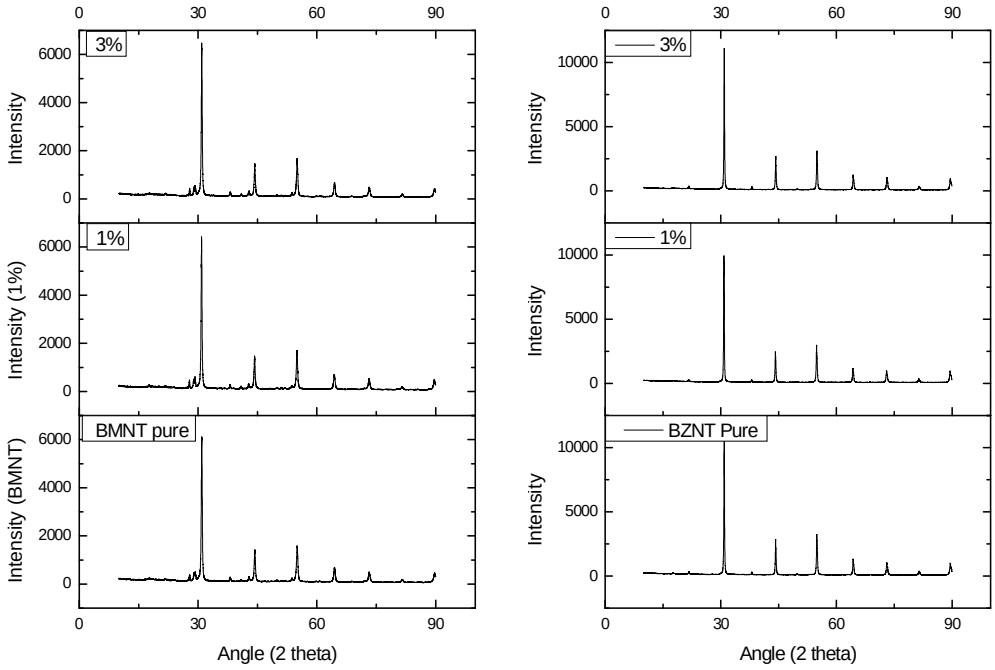
International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

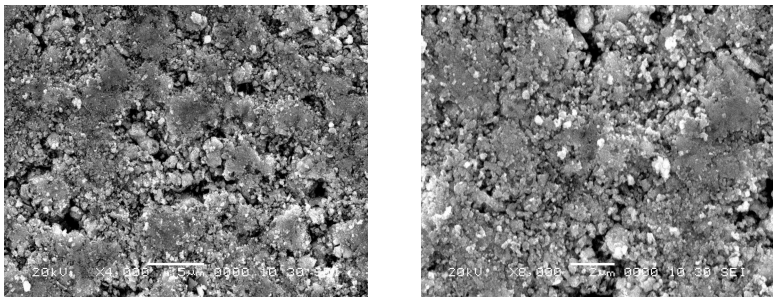
Vol. 3, Issue 6, June 2014



(c) **(d)**

Fig 1:XRD patterns of ceramic samples with and without B_2O_3

The SEM images are shown below:



BMN-pure

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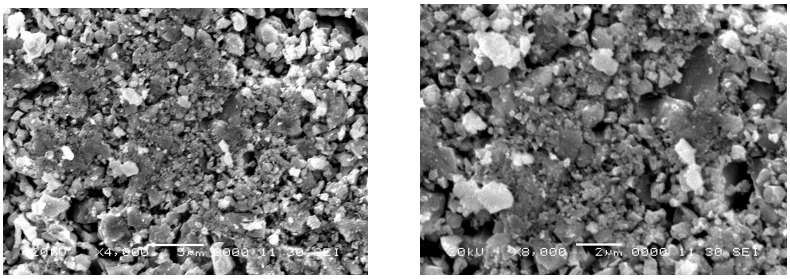
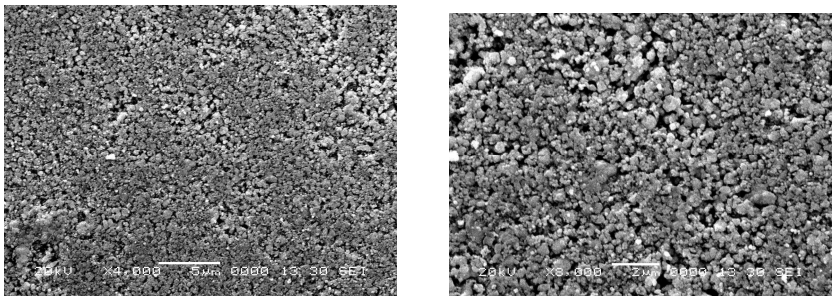
International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Vol. 3, Issue 6, June 2014



BZN-pure



BZNT-pure

Fig 2: SEM images of Pure BMN BZN and BZNT

III. RESULTS AND DISCUSSION

As discussed earlier, BMT gave better densification with the addition of frit glass. This can be seen from the table given below:

Composition	%wt loss after heating at 1300° C	Axial Shrinkage after heating at 1300° C	Radial Shrinkage after heating at 1300° C	Density in gm/ cm3 after heating at 1300°C	% theoretical density
BMT	1.557	13.88	0	4.38	57.7
BMT + 1% B_2O_3	1.346	12.30	10.89	6.44	84.4
BMT + 2% B_2O_3	0.894	12.12	12.53	6.23	82.2
BMT + 2.5% B_2O_3	0.690	13.49	12.5	7.39	97.36
BMT + 5% B_2O_3	3.278	No change	16.3	5.96	78.52
BMT + 7.5% B_2O_3	0.585	No change	4.69	4.23	55.7
BMT + 10% B_2O_3	0.888	0.77	No Change	3.87	50.98

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Vol. 3, Issue 6, June 2014

From the above densification data, after preparation of BMN, BZN, BMNT and BZNT, the samples were added with 1 to 3 % B_2O_3, and were pelletized and sintered at1250°C. The first heating cycle did not give proper densification of the samples. Hence, second heating cycle was carried out at same temperature. The results of these samples are given below:

Sample	Density (g/cm3)	Theoretical density	Percent density (converted)
BMN	4.327	6.236	69.39
BMN +1% B_2O_3	5.038	6.197	81.31
BMN+ 3% B_2O_3	4.572	6.121	74.70
BZN	5.117	6.511	78.59
BZN +1% B_2O_3	5.142	6.469	79.49
BZN+ 3% B_2O_3	5.297	6.387	82.93
BMNT	4.631	7.31	63.35
BMNT +1% B_2O_3	6.240	7.27	85.83
BMNT+ 3% B_2O_3	6.412	7.17	89.43
BZNT	5.302	7.57	70.04
BZNT +1% B_2O_3	6.827	7.52	90.78
BZNT +3% B_2O_3	6.638	7.42	89.46

From XRD patterns in fig.1, the desired perovskite phase formation can be seen at 2θ=30.9.The reported JCPDS no: 17-173 for BMN indicates the trigonal system and the JCPDS pattern no: 17-182 for BZN indicates the presence of cubic system. The JCPDS patterns for BMNT and BZNT are not reported.Thirumal*et.al* have reported that BMNT and BZNT sintered up to 1573 K consists of cubic phase [7]

But on comparison of BMNT with BMN, it isobserved that in both the samples, perovskite phase formation i.e. 100% peak is observed at 30.9°.Similarly, on comparison of BZNT with BZN, it can be observed that in both the samples, perovskite phase formation is seen at 2θ= 31°. XRD patterns of BMN and BMNT look iso-structural indicating that the crystal structure remains same but in case of BMNT and BZNT the B-site cation ordering is changing. Though there are large number of reports on synthesis, dielectric properties of BMT andBZT, but there is only one single report available for BMNT and BZNT [8].

From the SEM micrographs of BMN and BZN, it is observed that the structure has cavities which reduce the dielectric constant as well as mechanical strength of the ceramic. The image of BZN also shows presence of some secondary phase. As compared to BMN and BZN, BZNT shows better surface homogeneity. The SEM micrograph of BMNT is similar to that of its Zn analogue.

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Vol. 3, Issue 6, June 2014

Thirumal *et. al.* have reported that dielectric constant of BMNT decreases with increase in niobium concentration and varies from approximately 16 to 20. The dielectric constant for BMNT was found nearly in the desired range i.e. 15.64. The dielectric constant for all the ceramics deteriorates after the addition of B_2O_3. The constants are given in the table below:

Sample	Capacitance (PicoFarad)	Dielectric constant
1% BMN	2.67	2.025
3% BMN	4.03	2.459
1% BZN	6.48	3.398
3% BZN	5.62	3.459
BMNT	32.66	15.64
1% BMNT	3.97	2.07
3% BMNT	3.82	2.564
BZNT	12.55	5.947
1% BZNT	4.64	2.478
3% BZNT	4.85	2.317

IV. CONCLUSIONS

- Crystal structure of BMNT and BZNTwere found to be cubic.
- The maximum densification was achieved in different set of experiments as given below:

Sr. No.	Sample	Best Composition	% Density
1.	BMN	1% B_2O_3	81.31
2.	BZN	3% B_2O_3	82.93
3.	BMNT	1% B_2O_3	85.83
4.	BMNT	3% B_2O_3	89.43
5.	BZNT	1% B_2O_3	90.78
6.	BZNT	3% B_2O_3	89.46

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- On observing the dielectric constants achieved,it can be concluded thatin the present samples BMNT is having best dielectric const but addition of B_2O_3deteriorates its dielectric constant.Hence addition of B_2O_3 is not advisable in BMNT. So forachieving mechanical strength some other additive may be tried.
- Optimum sintering temp for BMNT has been reduced to 1250 i.e. by 235°C in comparison to that reported by Thirumal et.al.
- SEM images of BZNT indicate that better surface homogeneity has been achieved as compared to ternary parent compounds BZT and BZN.

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