

# **Systematic Study of Reaction Involved In Formation of Barium Magnesium Tantalate When Synthesized By Solid State Route**

N.Saraf<sup>1</sup>, R.V.Kashalkar<sup>2</sup>, M.Y.Khaladkar<sup>3</sup>, I.S.Mulla<sup>4</sup>, R. Purandare<sup>5</sup>.

PhD Student, Dept of Chemistry, S.P. College, Pune, Maharashtra, India<sup>1</sup>

Head, Dept of Chemistry, S.P. College, Pune, Maharashtra, India<sup>2</sup>

Associate Professor, Dept. of Applied Science, College of Engineering, Pune, Maharashtra, India<sup>3</sup>

CSIR Emeritus Scientist, Centre for Materials for Electronic Technology, Pune, Maharashtra, India<sup>4</sup>

PhD Student, Dept. of Applied Science, College of Engineering, Pune, Maharashtra, India<sup>5</sup>

**ABSTRACT:** Barium Magnesium Tantalate ( $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{2/3})\text{O}_3$ ) is  $\text{ABO}_3$  type perovskite material. It is fetching more attention of researchers as material for dielectric resonator and is used in satellite communication. The dominant mode of synthesis of this material is solid state route. In the present work TG/DTA has been carried out of the raw material in the temperature range from 30 to  $1400^\circ\text{C}$ . In TG 3 major weight-loss steps are observed till  $1000^\circ\text{C}$  with a total loss of 22% followed by weight gain of 18% in the temperature range 1000 to  $1300^\circ\text{C}$  and then weight loss continued till  $1400^\circ\text{C}$  and finally the stable composition showed a weight loss of 8%. This observation is supported by DTA which showed a broad endotherm till 200 suggesting loss of adsorbed water followed by a short endotherm at  $350^\circ\text{C}$ , a sharp endotherm at  $800^\circ\text{C}$  suggesting decomposition of Barium Carbonate. Enthalpy of the reaction mixture seems to rise quickly after the decomposition leading to ternary phase formation. The chemical composition changes rapidly after 1100 till  $1200^\circ\text{C}$  suggesting rapid uptake of oxygen. The sharp and significant endotherm observed between 1120 to  $1200^\circ\text{C}$  suggested that at  $1200^\circ\text{C}$  majority of the reactions taking place in the reaction mixture are over. Highest oxygen stoichiometry appeared at temperature  $1300^\circ\text{C}$ . Considering this, material was calcined at temperatures 950, 1200 2 hrs, 1200 18hrs, 1200 18 hrs (total 36 hrs) and 1300 2hrs. At every stage some amount of material was taken out and subjected for XRD. Crystallite size observed to be increasing from 200 to 331 Å at temperature 950 to  $1300^\circ\text{C}$  respectively.

**KEY WORDS:** BMT, TG/DTA, calcination, XRD, crystallite size.

## **I. INTRODUCTION**

An advance in wireless communication has become need of time. With advances in wireless technology one can get connected with any corner of the world in few seconds of time, one can even have seamless communication for long duration in any corner of world etc. This globalization is the outcome of extensive research in the miniaturization of materials which is getting used in the device. Barium magnesium tantalate ( $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{2/3})\text{O}_3$ ) is a very popular material for dielectric resonator. Low temperature, low cost and bulk quantity synthesis is still a challenge for researchers. This paper reports about the systematic study of reactions involved in synthesis of ( $\text{Ba}(\text{Mg}_{1/3}\text{Ta}_{2/3})\text{O}_3$ ) when synthesized by solid state route.

# International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Vol. 3, Issue 6, June 2014

## II . EXPERIMENTATION

For synthesis of  $(Ba(Mg_{1/3} Ta_{2/3})O_3)$  Sigma Aldrich  $Ta_2O_5$ ,  $MgO$  and  $BaCO_3$  were used. In that prior addition of stoichiometric amount of  $MgO$ , it was heated to  $1100^{\circ}C$  and furnace cooled to room temperature. These precursor materials were weighed in stoichiometric amount mixed in agate mortar for half hour rigorous grinding. Then this material ground in ball mill (at rate 100 rpm/min) using zirconia ball as grinding media in presence of iso-propyl alcohol (IPA) as a solvent for 2 hours. Then the material dried in oven and sieved. This raw material is now ready for characterization study and calcinations Raw material was subjected to TG/DTA in the temperature range ambient to  $1400^{\circ}C$ . Then, material is calcined at temperatures 950, 1200 2 hrs, 1200 18hrs, 1200 18 hrs (total 36 hrs) and  $1300^{\circ}C$  2hrs. At every stage some amount of material was taken out and subjected for XRD. A one gram material at  $1300^{\circ}C$  2 hrs was weighed, crushed and mixed with a drop of 5% Poly-vinyl alcohol (PVA) as binder in agate mortar. Then this compacted 10 mm puck using hydraulic pellet press at 5 tons pressure. Then this compact was subjected to heating from ambient to  $600^{\circ}C$  at the rate  $5^{\circ}C/min$  for two hour and furnace cooled. This pellet then was characterized by SEM and frequency response measurement.

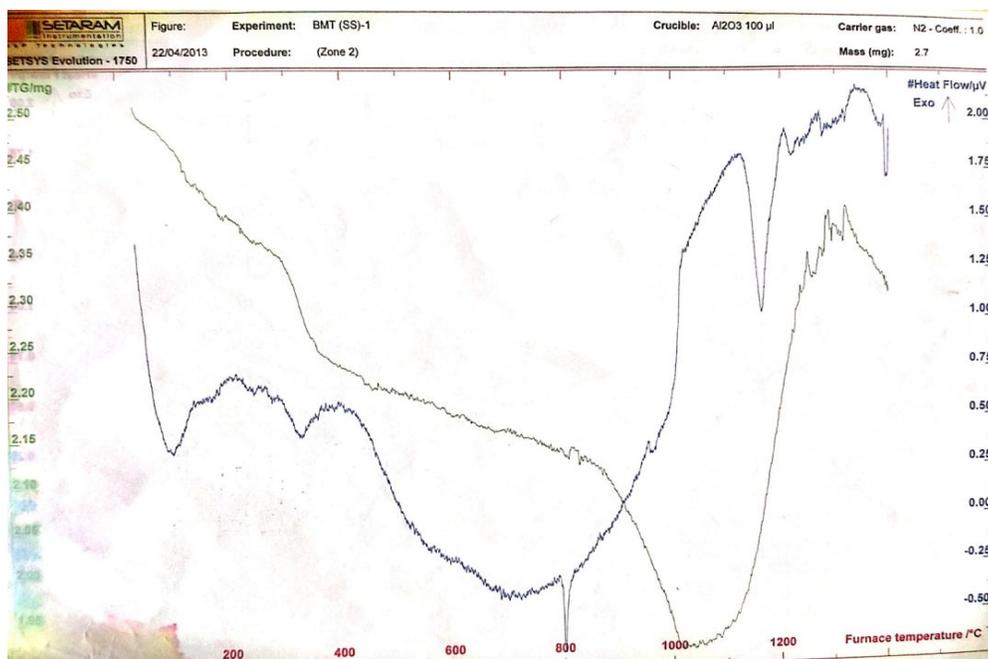


Fig. 1 TG/DTA plot of BMT when synthesized by solid state route.

# International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Vol. 3, Issue 6, June 2014

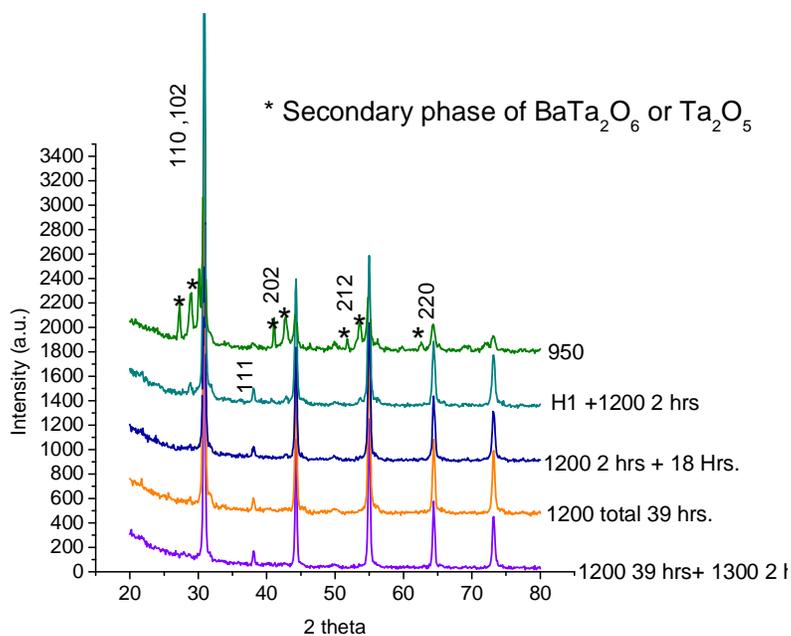


Fig.2 XRD pattern of BMT when calcined at different temperature.

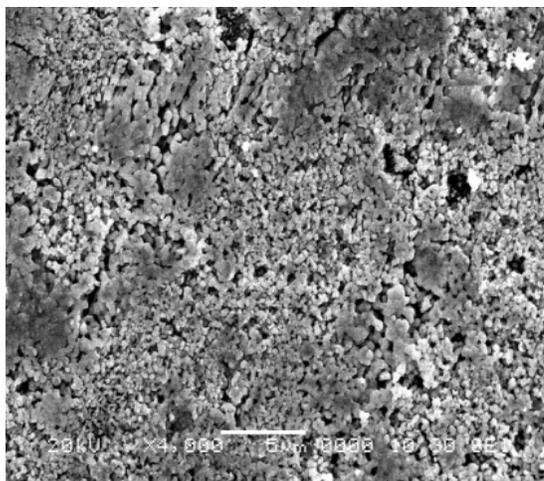


Fig.3 SEM image of BMT (SS) at 1300<sup>0</sup> C

# International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Vol. 3, Issue 6, June 2014

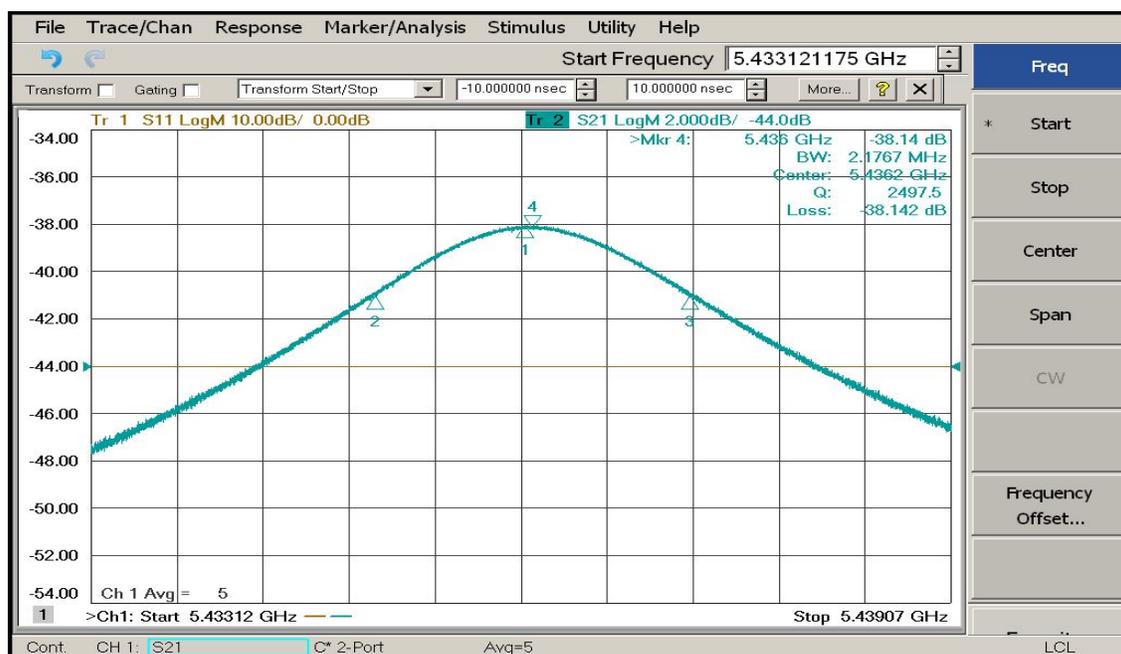


Fig.4: Frequency response measurement in the range 1 to 10 GHz.

### III RESULTS AND DISCUSSION

From Fig.1 TG weight-loss can be divided into three broad steps followed by a gain and slight loss . Following table gives idea about the temperature range for steps, percent weight-loss assigned for it and species lost.

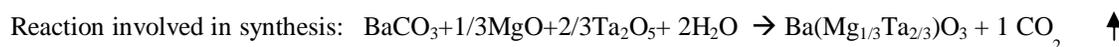
Table I  
Stepwise weight loss for TG graph.

Temperature range ( ° C )	% weight loss	Species lost
R.T to 250	6%	Adhered H <sub>2</sub> O
250 t 750	6%	2 H <sub>2</sub> O from BaCO <sub>3</sub>
750 to 1040	10%	1 CO <sub>2</sub> from BaCO <sub>3</sub>
1000to 1300	-18% wt (gain)	Oxygen uptake
1300 to 1400	Total loss of 8% from the start	Loss of oxygen
<b>Stable composition fits to Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub> at 1300</b>		

# International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Vol. 3, Issue 6, June 2014



In the present work TG/DTA has been carried out of the raw material in the temperature range from 30 to 1400°C. In TG 3 major weight-loss steps are observed till 1000°C with a total loss of 22% followed by weight gain of 18% in the temperature range 1000 to 1300°C and then weight loss continued till 1400°C and finally the stable composition showed a weight loss of 8%. This observation is supported by DTA which showed a broad endotherm till 200°C suggesting loss of adsorbed water followed by a short endotherm at 350°C, a sharp endotherm at 800°C suggesting decomposition of Barium Carbonate. Enthalpy of the reaction mixture seems to rise quickly after the decomposition leading to ternary phase formation. The chemical composition changes rapidly after 1100 till 1200°C suggesting rapid uptake of oxygen. The sharp and significant endotherm observed between 1120 to 1200°C suggested that at 1200°C majority of the reactions taking place in the reaction mixture are over. Highest oxygen stoichiometry appeared at temperature 1300°C. Considering this, sintering temperature was carefully chosen as 950, 1200- 2 hrs, 1200- 18hrs, 1200- 18 hrs (total 38 hrs) and 1300- 2hrs.

In XRD pattern (Fig.2) at 950°C shows presence of some secondary peaks which are either because of formation of binary BaTa<sub>2</sub>O<sub>6</sub> or because of some un-reacted Ta<sub>2</sub>O<sub>5</sub>. Further calcinations at higher temperatures show formation of single phase Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub>. At 1300°C characteristic peak of perovskite structure found to be developing at 2 theta angle 39. Table II shows crystallite size at different calcinations temperature.

From SEM image agglomeration observed with porosity. This observation shows that even if single phase forms at 1300°C for densification higher temperature is required or for densification at lower temperature additives or dopants are required. To study this observation authors have carried out effect of addition B<sub>2</sub>O<sub>3</sub> on physical characteristics and dielectrics of Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub><sup>[1]</sup>

Dielectric characterization of above synthesized sample showed Qxf 2497 at 5.433 GHz, loss factor - 38.14 and dielectric constant 22.4.

TABLE II

CRYSTALLITE SIZE CALCULATED AT DIFFERENT CALCINATIONS TEMPERATURES.

Sr. No	Heating History	C.S. (Å)
1	950	200.24
2	H-1 + 1200 2 hrs	275.82
3	1200 2 hrs + 18 hrs	289.37
4	1200 2 hrs+ 36 hrs	276.84
5	1200 36 hrs + 1300 2 hrs	331.59

# International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

Vol. 3, Issue 6, June 2014

## IV. CONCLUSIONS

- \* In TG stepwise weight-loss which is for loss of moisture, water of crystallization and loss of CO<sub>2</sub> observed till 1000<sup>0</sup>C.
- \* Weight gain is observed in the temperature range >1000 to 1200<sup>0</sup>C and continued till 1400<sup>0</sup>C which is for irregular uptake of oxygen making species oxygen rich.
- \* Crystallite size observed to be increasing from 200 to 331Å on heating the samples from 950 to 1300<sup>0</sup> C respectively.
- \* Dielectric characterization of above synthesized sample showed Qxf 2497 at 5.433 GHz, loss factor - 38.14 and dielectric constant 22.4.

## ACKNOWLEDGEMENT

Authors are thankful to ISRO-UoP STC cell University of Pune, for funding the work and Satellite Application Centre (SAC) Ahmedabad for analytical service support. We are also thankful to Department of Physics, Electronic Sciences University of Pune for analytical support.

## REFERENCES

1. Effect of B<sub>2</sub>O<sub>3</sub> glass additives on physical characteristics and dielectrics of barium magnesium tantalate Ba (Mg<sub>0.33</sub>Ta<sub>0.66</sub>)O<sub>3</sub>, N.Saraf ,R.Kashalkar, M.Khaladkar, I.Mulla, Int. J. app sci comp sci engg, (1) 2, 2012.
2. Effect of sintering process on microstructure characteristics of Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub> ceramics and their microwave dielectric properties, Mei-Hui Liang, Chen-Ti Hu, Hsiu-Fung Cheng, I-Nan Lin, John Steeds, Journal of the European Ceramic Society 21 (2001) 2759–2763.
3. Study on BMT millimeter wave dielectric resonators, Hongwei Chen, Chuanren Yang, Chunlin Fu, and Dayu Zhou, International Journal of Infrared and Millimeter Waves, 26, 2, February 2005.
4. Effect of preparation conditions on cation ordering and dielectric properties of Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub> ceramics, T.V. Kolodiazhnyia, A. Petrica, G.P. Johari, A.G. Belous, Journal of the European Ceramic Society 22 (2002) 2013–2021.
5. Dielectric loss and damping constants of lattice vibrations in Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub> ceramics, T. Shimada, Journal of the Euro. Ceram. Soc. 23 (2003) 2647–2651.
6. Study of second-phases in Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub> materials by microwave near-field microscopy, Hsiu-Fung Cheng, Yi-Chun Chen, Gang Wang, Xiao-Dong Xiang, Guan-Yu Chen, Kuo-Shung Liu, I-Nan Lin, Journal of the European Ceramic Society 23 (2003) 2667–2670.
7. Synthesis of Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub> microwave dielectrics by solid state processing, Yonghan Fang, Ang Hu, Yun Gu, Young-Jei Oh, Journal of the European Ceramic Society 23 (2003) 2497–2502.
8. Crystal structure of Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub> calcinated at 1400°C J.Srinivas, E D Dias, G Sreenivasamurthy, Bull. Mater. Sci., 20(1), (1997), 23- 25.
9. A new sol-gel route using inorganic salts for synthesizing barium magnesium tantalate ceramics, Chung-Hsin Lu, Chien-Cheng Tsai, Materials letters, 31 (1997), 271-274.
10. Effect of Ni substitution on the dielectric properties and lattice vibration of Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub>, T. Shimada, J. Euro.Ceram. Soc., 26, (2006), 1781–1785
11. Ba<sub>3</sub>ZnTa<sub>2.5</sub>Nb<sub>0.5</sub>O<sub>9</sub> and Ba<sub>3</sub>MgTa<sub>2.5</sub>Nb<sub>0.5</sub>O<sub>9</sub>: synthesis, structural and dielectric studies, M.Thirumal F, G. SenthilMurugan, K.B.R. Varmab, A.K. Ganguli, Mater Research Bull 35 (2000) 2423-2430
12. The effect of glass additives on the microwave dielectric properties of Ba(Mg<sub>1/3</sub>Ta<sub>2/3</sub>)O<sub>3</sub> ceramics, K.P. Surendran, P. Mohanan, M.T. Sebastian, J. Solid State Chem., 177 (2004), 4031–4046