Effect of tantalum substitution on dielectric constant of $ZnSb_{2-x}Ta_xO_6$ solid solution (x = 0.0, 0.1, 0.25, 0.75, 1.6)

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Antimonates and tantalates of transition metals of the general formula MM'_2O_6 (M = Zn, Cd, Pb, Ni; M' = Sb, Ta) are still being investigated due to their interesting structural, electrical and optical properties [1–3]. Antimonates MSb_2O_6 (M = Zn, Mg, Ba, Cd) are *n*-type semiconductors [1–3]. ZnSb₂O₆ and MgSb₂O₆ make interesting electrode material for dye sensitized solar cells–DSSC [1]. Recently, much attention has been paid to ZnSb₂O₆ and ZnTa₂O₆ because of their possible application in photocatalysis and sensors for detection of nitrogen oxides and hydrogen sulfide [4] as well as materials for dielectric microwave devices used for satellite communication [5]. In the ZnSb₂O₆–ZnTa₂O₆ system a limited substitutional solid solution of the formula ZnSb_{2-x}Ta_xO₆ for $0 < x \le 1.6$ crystallized in the tetragonal adopting the tri-rutile type structure. The IR and UV-vis-NIR studies showed the extension in the crystal lattice and the shift in the position of IR absorption bands toward higher wave numbers as well as the increase of energy gap in the range 3.65-4.60 eV for $0.25 \le x \le 1.60$ [6].

The electrical conductivity was measured by the DC method using a KEITHLEY 6517B Electrometer/High Resistance Meter. Broadband dielectric spectroscopy measurements were carried out using pellets, polished and sputtered with (~80 nm) Ag electrodes in the frequency range from 200 Hz to 5 MHz with a LCR HITESTER (HIOKI 3532-50, Japan) within the temperature range of 79–400 K.

The solid solution of ZnSb_{2-x}Ta_xO₆ (x = 0.0, 0.1, 0.25, 0.75, 1.6) is insulators at low temperatures and slowly activated at higher temperatures. Broadband dielectric spectroscopy measured in the temperature range of 79- 400 K showed a strong dependence on temperature and frequency both dielectric constant, ε_r , and loss tangent, tan δ , for poorer samples in tantalum and poor dependence on temperature and frequency for rich samples in tantalum. The electrical relative permittivity, ε_r , reaches a value up to 1,500 for samples that are poorer in tantalum, *i.e.* in the content range of $0 \le x \le 0.25$. These samples show high losses above tan $\delta > 10$. Whereas samples richer in tantalum, *i.e.* for x = 0.75 and 1.6, have low value of $\varepsilon_r \sim 10$ and very low losses below tan $\delta = 0.01$. The most spectacular result of this research is a dramatic change in ε_r and tan δ caused by tantalum doping for x = 0.75. The above results are interpreted as part of the dipole relaxation for the poorer samples in tantalum and as the spatial charge polarization for samples richer in tantalum, for which the freedom of electron or ion charge is limited.

[1] J. Jang, and S.-J. Kim, Jpn. J. Appl. Phys. 51, 10 (2012).

- [2] A. Katsui, K. Hatano, and H. Matsushita, J. Jpn. Soc. Powder Netall. 54, 479 (2007).
- [3] N. Kikuchi, H. Hosono, H. Kawazoe, O. Tanegashima, I. Ota, and Y. Kimura, J. Am. Ceram. Soc. 88, 2793 (2005)
- [4] W. Liu, P. Lin, H. Xue, Y. Zhang, and Z. Li, J. Mol. Catal. A 349, 80 (2011).
- [5] H.J. Lee, I.T. Kim, and K.S. Hong, J. Appl. Phys. 36, 1318 (1997).
- [6] G. Dąbrowska, *Therm. Acta* **614**, 62 (2015).