BEHAVIOR OF THE MAIN PROPERTIES OF HARD AND SOFT TYPE PIEZOCERAMICS WITH TEMPERATURE FROM 2 TO 300 K

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1. INTRODUCTION

PZT materials, consist of solid solutions between lead titanate and lead zirconate, on a relatively narrow interval of compositions, around the composition with the formula $Pb(Zr_{0,5}Ti_{0,5})O_3$, situated within the morphotropic zone boundary, where two types of crystalline structures coexist, one tetragonal and one rhombohedral, and where the piezoelectric properties have unusual maxima. Their basic property is the so called direct and converse piezoelectric effect, by which an electrical charge (a voltage) is generated when it is mechanically stressed (direct effect) and vice versa a mechanical deformation is developed when subjected to an electrical field (converse effect). In applications, these materials work at room temperatures. But there are also many applications where they have to work at other temperatures, above the ambient so that the requirements for such materials is to have constant properties over a large temperature interval.

2.OBJECTIVES

- To prepare two piezoelectric materials of high performance based on PZT system;
- The chemical formula of the soft material was:

  $PbNb_{0.02}Li_{0.007}Zr_{0.51}Ti_{0.463}O_3$;

and the hard material:

  $PbMn_{0.017}Sb_{0.033}Zr_{0.48}Ti_{0.47}O_3$;

- To investigate the temperature dependence of the main piezoelectric parameters within a wide temperature interval from 2 to 600 K;
- To determine the maximum temperature up to which the materials can be safely used for transducers.

3. EXPERIMENTAL

The materials chosen for the experiment were a typically soft and a typically hard PZT material. The preparation method was the conventional mixed oxide route, using p.a. purity raw oxides. The stoichiometric amounts of oxides were wet mixed for 2 hours in a planetary ball mill using agate vessels and balls of 10 mm diameter at a ball/oxide powder ratio of 3/1. The mixed powders were calcined at 900 ^oC for 3 h followed by milling for 6 h. The calcined powders were checked by X-ray and the results showed only the perovskite single phase for both materials. Disc shaped samples of 12 mm diameter and 1.5 mm thick were next uniaxially pressed and sintered for 3 hours at 1250 ^oC (soft material) and 1300 ^oC (hard material) respectively. At these temperatures the materials were fully densified showing average densities of 7.72 and 7.70 g/cm^3 for the soft and hard material respectively, which represent more than 97,5 % of TD. Metallographic examination of polished surfaces of sintered samples showed a well uniform and dense structure with mean grain size of about 0.6-0.7 µm. After mechanical processing the samples were electroded with a thin layer of Ni chemically deposed and poled in a silicon oil bath at 220 ^oC under an electric field of 30 kV/m. The piezoelectric measurements were carried out after 48 hours of relaxation at room temperature.

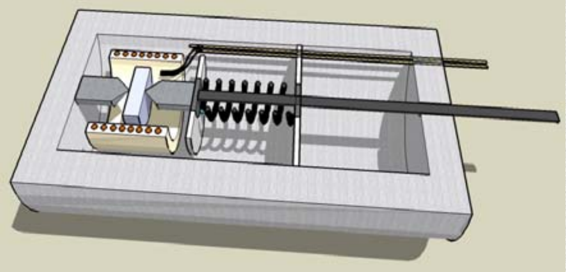

***Fig. 1.* Schematic view of the temperature chamber for measurements above room temperature**

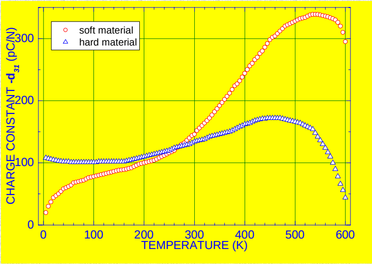

Fig. 5. *Temperature dependence of the charge constant – d_{31} for soft and hard materials*

4. RESULTS

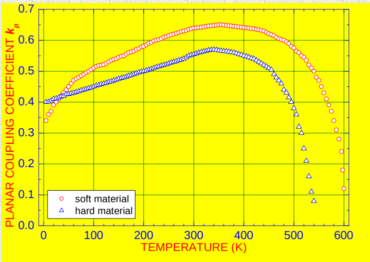

Fig. 2. *Temperature dependence of the planar coupling coefficient k_p for soft and hard materials*

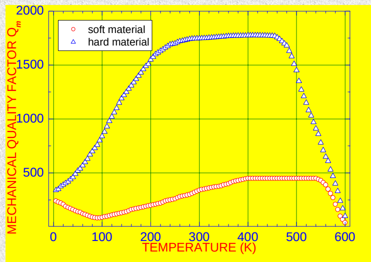

Fig. 3. *Temperature dependence of the mechanical quality factor Q_m for soft and hard materials*

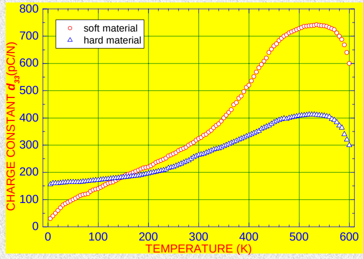

Fig. 4. *Temperature dependence of the charge constant d_{33} for soft and hard materials*

A number of piezoelectric constants k_p, Q_m, d_{33} and d_{31}, g_{33} *and* g_{31}, ε_r, were determined as a function of temperature:The behavior of the coupling coefficient k_p with temperature is shown in figure 2. It can be seen that k_p for the hard material increases steadily from 0.4 at 2K to about 0.57 at 350 K then drops drastically by approaching the Curie temperature. A similar behavior is also recorded for the soft material except that the initial increase between 2 and 80 K is much more pronounced after which the increase becomes continuously steady up to about 450 K.

Figure 3 shows the behavior of the mechanical quality factors Q_m vs temperature. For the hard material it increases drastically by about 500 % between 2 and 250 K, then very slowly and steadily up to 460 K after which it suddenly drops. The behavior of Q_m for the soft material is quite different. At first, between 2 and 80 K, it decreases more than twofold then increases linearly by 350% up to 400 K, remains constant up to nearby Curie point and then suddenly drops to zero.

Charge constants d_{33} and d_{31} exhibit a similar behavior namely a steady increase with increasing temperature as it results from figures 4 and 5 respectively, but at different values. If for the hard material d_{33} increases about threefold between 2 and 550 K, the increase for the soft one is thirty fold greater. The corresponding figures recorded for the increase of d_{31} were 1.7 times for the hard and 10 times for the soft one respectively.

On the same line the voltage constants g_{33} and g_{31} show a steady and continuous decrease with temperature up to the vicinity of the Curie point, where they start to drop much more rapidly, as can be seen in figures 6 and 7.

Finally, figure 8, illustrates the dependence of the relative dielectric constant ε_r on temperature. As one can see there is no noticeable difference for the two materials at least up to 450 K. The dielectric constants increase rather constantly from 200 at 2 K up to about 3600 around 470 K, which represents an increase by 18 times. From this temperature on the dielectric constants increase at much higher rates, reaching maximum values of 15000 and 20000 for the hard and soft materials respectively, at their Curie points of 520 K and 620 K respectively.

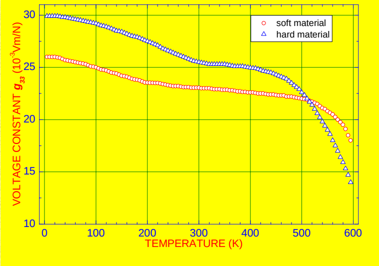

Fig. 6. *Temperature dependence of the voltage constant g_{33} for soft and hard materials*

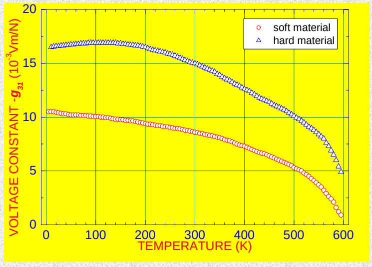

Fig. 7. *Temperature dependence of the voltage constant $-g_{31}$ for soft and hard materials*

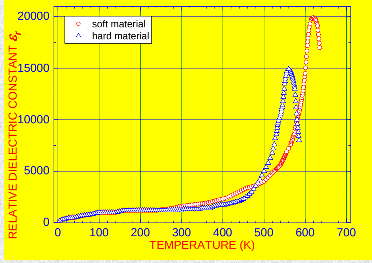

Fig. 8. *Temperature dependence of the relative dielectric constant ε_r for soft and hard materials*

5. SUMMARY

A general examination of the experimental results obtained on the two PZT materials investigated reveals the following aspects: The planar coupling coefficients show a continuous increase with increasing temperature, having maximum values around room temperature; The soft material has the tendency to become a little harder with increasing temperature, and the hard one to become strongly harder showing a plateau of the highest values of Q_m for temperatures just a little above room temperature; Piezoelectric charge constants d_{33} and d_{31} increase steadily with temperature showing humps with the highest values at temperatures around 540 K; Piezoelectric voltage constants g_{33} and g_{31} decreases constantly with increasing temperature up to temperatures in the vicinity of the Curie point; High dielectric activity was recorded for both materials with a constant increase up to 450 K and a drastic increase above it showing maximum values at the Curie point.