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Effect of Temperature on The Main Piezoelectric Parameters of A Soft PZT Ceramic

C. MICLEA[1], C. TĂNĂSOIU[1], L. AMARANDE[1], C.F. MICLEA[1], C. PLĂVIŢU[2], M. CIOANGHER[1], L. TRUPINA[1], C.T. MICLEA[2], C. DAVID[2]

[1]National Institute for Materials Physics, Bucharest-Măgurele, România
[2]Hyperion University, Calea Călăraşilor 169, sector 3, Bucharest, România
E-mail: cmic@infim.ro

Abstract. The temperature behavior of the main piezoelectric parameters, such as the electromechanical coupling factor k_p, the mechanical quality factor Q_m, the dielectric permittivity ε (real and imaginary part), the loss tangent $\tan\delta$, the piezoelectric charge constants d_{33} and d_{31} and the voltage constants g_{33} and g_{31} of a soft type piezoelectric material was investigated. The results showed that temperatures under 150°C do not influence these parameters, which indicate that every transducer made from this type of material may be successfully used up to this temperature. Between 150°C and 250°C, the piezoelectric properties undergo more or less important changes, mainly due to the depoling effect and above that, at temperatures over 250°C, they degrade very rapidly, tending to zero.

1. Introduction

The piezoelectric effect was discovered by the end of the last century [1–4] and it consists in that certain materials when subjected to mechanical stress become electrically charged at their surface and vice versa. This effect remained rather a curiosity until the early 1920 when it was utilized to realize crystal resonators to stabilize the oscillations and thereby launching the field of frequency control. Anyhow, the piezoelectric materials have been studied more intensely only by the early 60s, after Jaffe's discovery of a new class of piezoceramic materials [5] namely the lead titanate-zirconate system with better properties than those known until then (quartz, Rochelle salt and barium titanate) [6–8]. This category of materials, called PZT materials, consists of solid solutions between lead titanate and lead zirconate, within a relatively narrow interval of compositions, around $Pb(Zr_{0,5}Ti_{0,5})O_3$, a composition situated within a zone called morphotropic zone boundary, where two types of crystalline structures coexist, one tetragonal and one rhombohedral, and where the piezoelectric properties have unusual maxima [5], which makes them very useful for applications in almost all areas of activity [9–12]. The basic properties of these materials are the so called direct and converse piezoelectric effect consisting in the generation of an electrical charge (a voltage) when it is mechanically stressed (direct effect) and vice versa developing a mechanical deformation when subjected to an electrical field (converse effect). In short piezoelectricity is defined as generation of electricity as a result of mechanical pressure or the phenomenon in which an electric polarization is produced by mechanical strain in materials belonging to a certain class of crystals, without a centre of symmetry.

In general, in applications, these materials work at room temperatures or on a relatively narrow temperatures interval around the ambient. But there are also many applications where they have to work at temperatures above the ambient. Therefore, it becomes compulsory to know the behavior of their main parameters with temperature in order to make the proper design for any specific application.

This work investigates the behavior of the main parameters of such materials up to the Curie temperature, where they become paraelectric and, therefore, inactive from the piezoelectric point of view.

2. Experimental

2.1. Material

The chemical formula of the soft material investigated was: $PbNb_{0.02}Li_{0.007}(Zr_{0.51}Ti_{0.463})O_3$. It was prepared by the usual ceramic method, consisting in mixing p.a. purity oxides followed by solid state reaction. The mixture was made in a planetary ball mill, in agate jars, in methanol, for 2 hours. The slurry was dried by continuous stirring, in order to avoid the gravimetric separation of the heavy elements. It was then double calcined at 850°C and 900°C, for 2 hours. The calcined product was now milled in the same mill, for 6 hours. After this, the powder was pressed in a steel die, at a pressure of about 50 MPa, as discs with the diameter of 12 mm and the thickness of 1.5 mm and as parallelipipeds, with dimensions of $60\times14\times14$ mm^3, and sintered in alumina crucibles, at a temperature of 1 270°C, for 6 hours.

The sintered samples were mechanically processed as discs with plane parallel faces having the diameter of 10 mm and the thickness of 1 mm, and as parallelipipeds of $20\times5\times5$ mm^3 using some special cutting and polishing machines. These dimensions are standard ones, required to determine correctly the material parameters [13].

Ni electrodes were applied on both faces of the discs and on the tops of the parallelepiped samples and then, they were poled in an electric field of 3 kV/mm, in a silicon oil bath, at the temperature of 220°C. After poling and relaxation (at least 24 hours), the material parameters were determined.

2.2. Measurement

In order to determine temperature behavior of the material's characteristics, a special device was built, and adapted to the measuring instrument used, namely to the impedance analyzer Agilent 4294A. The measurement method was the resonance-antiresonance technique. An automatic data acquisition and processing program was developed and the instrument was connected to a computer through a proper interface.

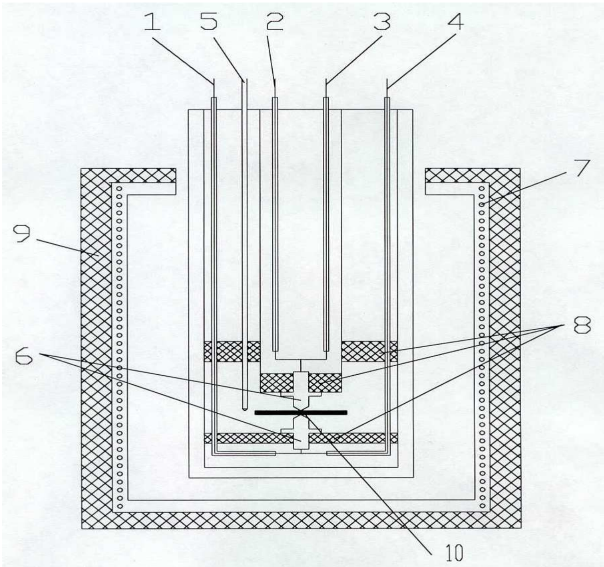

Fig. 1. Experimental setup for measuring the temperature behavior of the piezoelectric parameters.

The measuring device is shown in Fig. 1. It consists in the following main elements: **1, 2, 3** and **4** are the coaxial cables for signal that resist at high temperatures; **5** the chromium-nickel thermocouple placed near the sample; **10** the sample to be measured; **6** pines for holding the sample, which also assure the electrical connection; **7** heating resistance; **8** isolating ceramic; **9** thermo-insulating outer case.

For the calculation of the material parameters there were used the formulas furnished by the latest IEEE standards [13], on samples having the shape and dimensions required by these standards in order to have negligible measurement errors.

3. Results and discussions

The piezoelectric constants determined in this experiment are the following:

– the electromechanical coupling factor, k_p;

– the mechanical quality factor, Q_m;

– the relative dielectric constant, ε_r;

– the loss tangent, $\tan\delta$;

– the charge constants, d_{33} and d_{31};

– the hydrostatic piezoelectric coefficient, d_h;

– the voltage constants, g_{33} and g_{31}.

Figure 2 shows the variation of the electromechanical coupling factor k_p with temperature, from room temperature up to the Curie point, situated around 350°C.

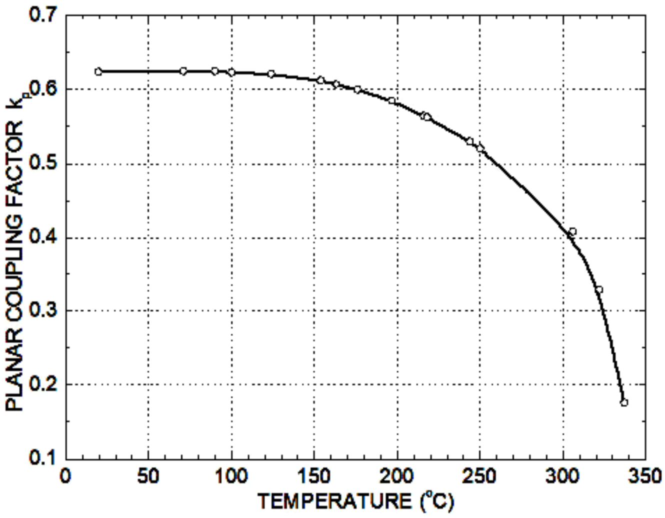

Fig. 2. The temperature dependence of the electromechanical planar coupling coefficient k_p between room temperature and the Curie point.

One observes that up to 150°C k_p remains constant after which it decreases relatively slowly up to about between 250°C, with a decreasing rate of about 0.2%/°C and above 250°C, the decreasing is rather sudden. At 350°C, the coupling coefficient has a value of just 0.1.

This behavior may be understood in terms of the mobility phenomena of ferroelectric domains walls. Until 150°C, the walls are moving insignificantly and so the sample remains practically in the same poling condition as at room temperature. Between 150°C and 250°C, the depoling process takes place rather slowly. Over 250°C, the depoling takes place more rapidly, and the piezoelectric properties decrease in the same manner, tending to zero. What it is most remarkable is the fact that the material doesn't change its coupling factor up to rather high temperatures (for example at 150°C), which makes possible that transducers made with this material to be used, with great efficiency, even at high temperatures.

Figure 3 illustrates the behavior of mechanical quality factor Q_m with temperature. One can also observe that between the room temperature and 250°C it grows up slowly and steadily after which it remains nearly constant up to about 300°C and then suddenly falls off due the quick depoling of the sample.

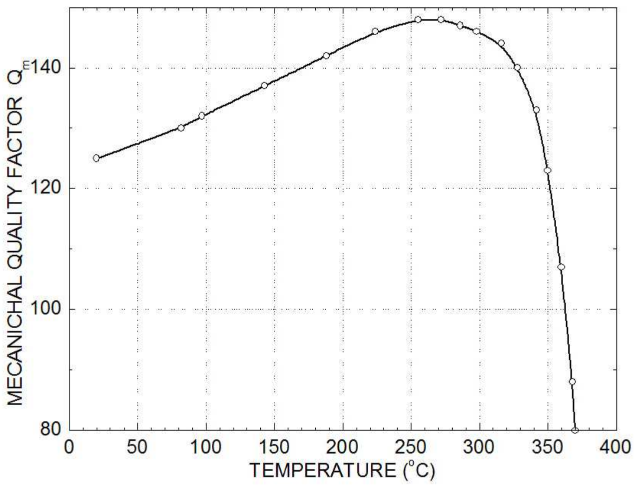

Fig. 3. The temperature behavior of the mechanical quality factor Q_m between room temperature and the Curie point.

Figure 4 presents the behavior of the dielectric constant with temperature of the real part as well as of the imaginary one too. In the case of this parameter, one observes an insignificant increase of the real part up to 250°C, after which it suddenly goes up, achieving a maximum of almost 20 000 around Curie temperature. The imaginary part remains constant over the whole temperature interval presenting only an insignificant maximum around Curie temperature, which seems normal, having into consideration the direct relation between these parameters.

The behavior of the loss tangent $\tan \delta$ is shown in Fig. 5. One can see that it behaves similarly to the dielectric constant which seems normal taking into consideration their direct connections.

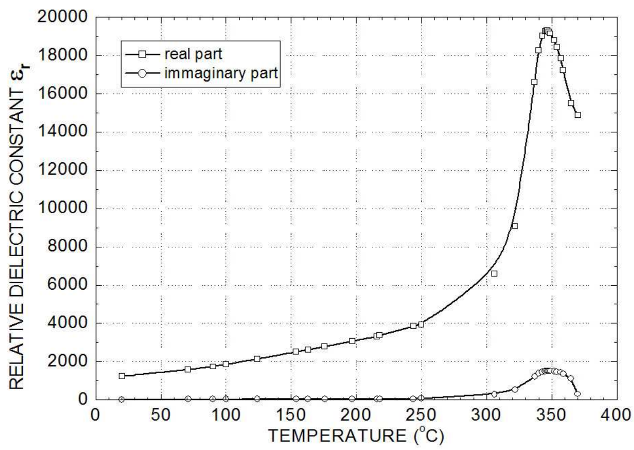

Fig. 4. The temperature dependence of the dielectric constant ε between room temperature and the Curie point.

It remains practically constant up to 250°C at a value of about $2{\cdot}10^{-3}$ and then suddenly increases to the high value of $8{\cdot}10^{-3}$.

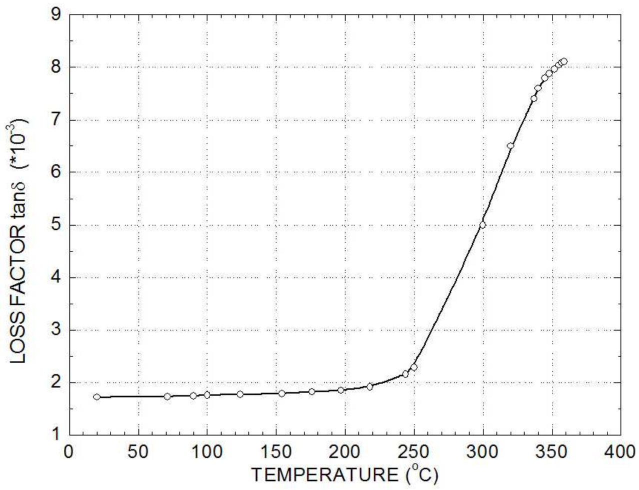

Fig. 5. The temperature dependence of the loss tangent $\tan\delta$ between room temperature and the Curie point.

The behavior of the charge constants d_{33} and d_{31} are presented in Fig. 6 while the hydrostatic piezoelectric constant d_h is shown in Fig. 7.

The relation between these three constants is given by the relation: $d_h = d_{33} + 2(-d_{31})$. The coefficient d_h is very important when construction of sonars is involved. The quality of these sonars is given by the great value of d_h and by its constant dependence in relation with temperature.

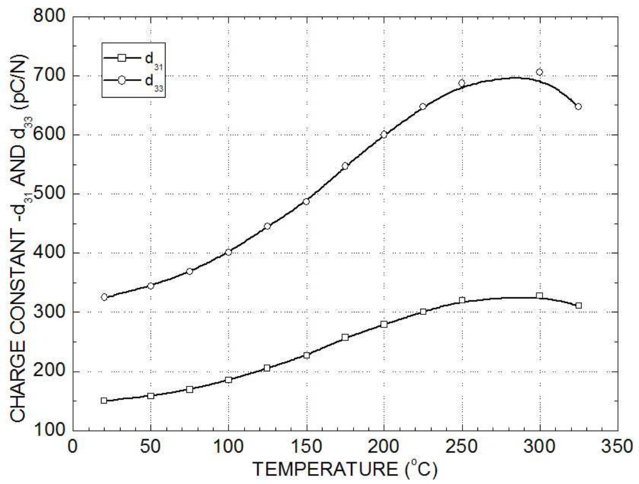

Fig. 6. The temperature dependence of the charge constants d_{33} and d_{31} between room temperature and the Curie point.

In Figure 7 it is observed that on the entire temperature interval from room up to the Curie point T_C, it increases steadily with a rate of only 0.1 pC/N which could be considered as practically constant. This makes that the discussed material to be very promising for the realization of this type of devices.

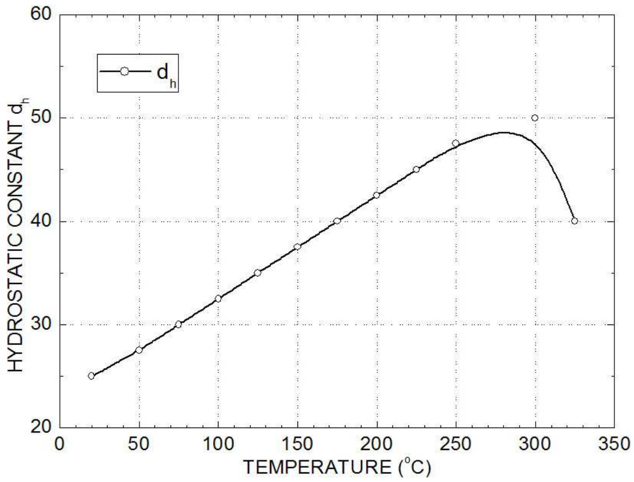

Fig. 7. The temperature dependence of the hydrostatic coefficient d_h between room temperature and the Curie point.

Figure 8 illustrates the temperature dependence of the voltage constants $-g_{31}$ and g_{33}. One observes that g_{33} remains practically constant up to 250°C and then decreases rather suddenly while g_{31} shows a rather steady decrease with a rate of about 0.03 Vm/N/°C over the whole temperature interval.

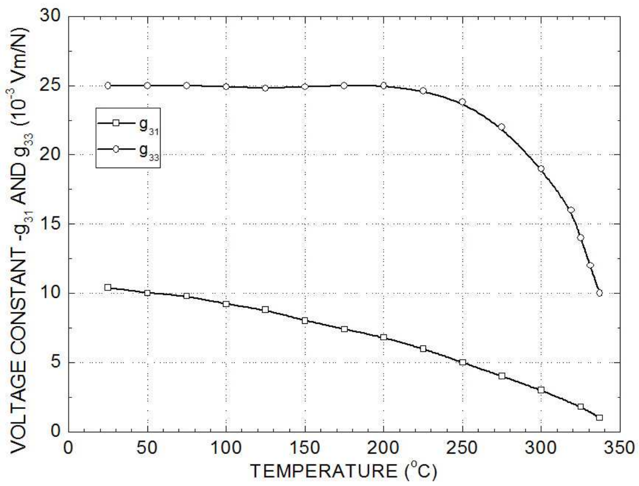

Fig. 8. The temperature dependence of the voltage constants g_{33} and $-g_{31}$ between room temperature and the Curie point.

4. Conclusions

The variation of the main piezoelectric parameters of a soft type PZT material was studied as a function of temperature on a large temperature interval, from room temperature up to the Curie point.

From this study it was concluded that all these parameters do not present significant variations up to the temperature of 150°C which makes that the transducers made with this material to be performant up to this relatively high temperatures. Between 150°C and 250°C, their performances decrease slightly but not so as to be essentially affected. Over 250°C, the transducers cannot be used since the piezoelectric parameters drastically and irreversibly decrease.

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