High temperature stability of new single crystal piezoelectric sensors

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Abstract

A new generation of high temperature piezoelectric sensors offering new perspectives in terms of performance and availability is in development. The sensors are based on superior synthetic monocrystalline piezoelectric materials. These crystals, known under the tradename PiezoStar® have been proved in numerous applications in harsh environments since many years. Among the outstanding properties of these crystals is their ability to perform continuously at temperatures above 600°C, the absence of pyroelectricity, great stability with no twinning and no phase transition up to the melting point (above 1300°C) as well as high sensitivity and mechanical strength. This paper examines the critical properties of PiezoStar® sensing elements for high temperature dynamic pressure sensors and accelerometers and compares them against known materials. The stability of the electrical insulation resistivity in different atmospheres has been investigated as function of temperature and time. In addition to the investigation of the PiezoStar® crystals several types of prototype sensors were built and initial tests conducted. Results available thus far indicate that PiezoStar® crystals are well suited for high temperature pressure sensors up to 600°C for continued operation and around 700°C for intermittent operation.

1. Introduction

The demand for sensors, especially pressure and vibration sensors, operating in harsh environments with high reliability at high temperatures is mainly driven by requirements regarding safety, emissions and costs. Typical applications include power generation, combustion engines and turbomachinery. Piezoelectric pressure sensors or accelerometers are widely used when it comes to accurately measure the pressure in the combustion chamber of engines or monitoring structures submitted to high vibrations. The quality of the sensors is largely defined by the high temperature performance and long term stability of sensing elements. Existing piezoelectric sensors operating above 450°C are based on natural or artificial piezoelectric materials, such as tourmaline crystals or special piezoceramics. In the last two decades new crystals, such as gallium orthophosphate ($GaPO_4$) or langasite ($La_3Ga_5SiO_{14}$) [1, 2], were used in sensors. Langasite-type crystals belong to the family of $Ca_3Ga_2Ge_4O_{14}$ (CGG) crystals, which are reported to be potential candidates for high temperature sensor applications. Different compounds of this family have already been grown and characterized. However, all sensing elements in use today have certain drawbacks either with respect to pyroelectricity, stability at high temperatures, excessive anisotropy or limited availability.

In this work a new class of piezoelectric single crystals, known under the trade name PiezoStar® will be presented. These crystals exhibit very promising properties for high temperature applications above 450°C and appear to overcome several of the drawbacks commonly seen in sensing materials used today. Up to now, these new crystals are used extensively in pressure sensors for combustion engines. For operation at temperatures up to 600°C new pressure and acceleration sensors were designed, manufactured and tested. Special care has been taken to achieve high long term stability of the measuring properties.

2. Requirements for a high temperature piezoelectric measurement chain

The typical piezoelectric measurement chain consists of a piezoelectric sensor connected to a charge amplifier with a high impedance cable and a data acquisition unit. High temperature applications have additional requirements due to harsh environment:

i) The sensors have to be ground insulated to minimize electromagnetic interferences. Hence differential charge amplifiers have to be used.
ii) Hardline mineral insulating cables must be used when the temperature exceed 400°C.
iii) The sensors are mainly used for monitoring applications and they should have a very long lifetime (10’000 h, possibly 30’000 h)

The piezoelectric measuring technique requires sensors with high electrical insulation. The main reason is that leakage currents due to the offset voltage at the input stage of the charge amplifier induce a drift current which leads to saturation of the charge amplifier. Saturation can be avoided with a high-pass filter, but the lower frequency limit increases as the resistance of the sensor decreases. As the resistivity of insulators decreases at high temperature, the maximal operating temperature of a sensor in a given frequency range is actually limited by the insulation resistance of the sensor (piezoelectric crystal).

3. Requirements for high temperature piezoelectric materials

Above 400°C, the choice of available piezoelectric material narrows dramatically. On the one hand, the insulation resistance of every material drops as the temperature increases; on the other hand the stability of the physical material properties (insulation resistance, piezoelectric sensitivity) becomes a serious issue, limiting the lifetime of the sensor.

As previously mentioned, high electrically insulating materials are needed to prevent the lower frequency limit of the measuring chain to increase above the frequency of interest. Only a few materials are known to have very high insulation properties, among them quartz, tourmaline, gallium orthophosphate and the new PiezoStar® crystal KI100. Other piezoelectric materials like piezoceramics, langasite, langatate or lithium niobate have poor insulation properties.

The stability issue is related to the degradation of some physical properties of the materials at high temperature. For instance, ferroelectric materials tend to lose their polarization as a result of domain switching. Thus, the piezoelectric sensitivity decreases with time, the quicker the higher the temperature. Piezoceramics are poled during the fabrication process, usually at temperature around 200°C, and depolarization occurs when the temperature rises above the poling temperature, even if the Curie temperature is much higher than the operating temperature. The stability issue of the sensitivity can neither the less be avoided or at least strongly reduced by using non ferroelectric crystals like those belonging to the crystal class 32. In addition, these crystals should not undergo any phase transition up to the melting point; otherwise twinning may occur, which in turn leads to an irreversible decrease of the sensitivity. Both quartz and gallium orthophosphate are prone to twinning, hence limiting their use at temperatures much lower than the transition temperature.

Besides sensitivity, resistivity may also show stability issues. Most of the piezoelectric sensing elements are oxides and oxides are known to be unstable at high temperature in a reducing atmosphere. The oxidation state at the surface of the piezoelectric crystal is reduced as weakly bonded oxygen atoms are removed from the surface. As a result, the surface resistivity decreases. The decrease rate depends on the temperature, the oxygen partial pressure and the strength of the oxygen bond. The effect is usually observed at temperatures above 400°C and when the oxygen partial pressure decreases below 10^{-4} mbar. Note that in air, the resistivity of oxides is stable. Unfortunately, the sensors are usually hermetically sealed in vacuum to prevent any corrosion to occur within the sensor.

The piezoelectric material should not respond to temperature changes. This is particularly important when measuring small signals like pressure fluctuations. For this reason, the use of non-pyroelectric materials is to be favored.

4. PiezoStar® KI100: a new high temperature piezoelectric crystal

The PiezoStar® crystals are a family of piezoelectric crystals for high temperature applications that have been developed by Kistler in collaboration with the Leibniz Institute of Crystal Growth (IKZ, Berlin, Germany) during the last 15 years. Their structure is similar to the calcium gallogermanates ($Ca_3Ga_2Ge_4O_{14}$) [2], like langasite or langatate, and they belong to the crystal class 32. They are grown from the melt according to the Czochralski process [3] and it takes about 2 weeks to growth a 2 kg crystal, with a diameter of 60 mm and a length of 180 mm (Fig. 1). This is a rather quick way to obtain crystals with good quality, compared to quartz (3 to 6 months) or gallium orthophosphate (12 months) with the hydrothermal growth.

For sensors, Kistler is using different crystals from the PiezoStar® family besides quartz. KI100 is the high temperature crystal par excellence. Its sensitivity is double that of quartz, it has temperature compensated cuts and it is stable at least up to 700°C. KI85 has a high sensitivity (4 to 5 times that of quartz) and is used in applications where the temperature does not exceed 500°C. Note that none of these crystals are pyroelectric (unlike tourmaline, lithium niobate or piezoceramics) and that they are twinning free, as they do not undergo any phase transition up to the melting point. The properties of these crystals are shown in the chapter 6 and a comparison with other high temperature materials is given in Table 1.

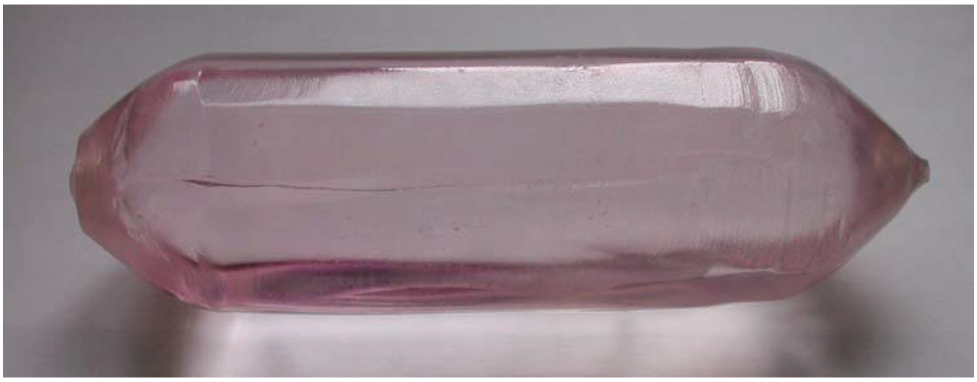

Fig. 1: PiezoStar® crystal KI100, diameter 60 mm, length 180 mm, weight 2 kg (courtesy of Leibniz Institute of Crystal Growth).

	T max °C	sensitivity pC/N	resistivity	twinning	pyro-electricity	crystal class	growth
KI100		5	high	no	no	32	Czochr.
Tourmaline	700	1.7	high	no	yes	3m	natural
$GaPO_4$		4.5	high	yes	no	32	hydroth.
KI85		9.1	medium	no	no	32	Czochr.
Langasite	500	6.2	low	no	no	32	Czochr.
Langatate		6.9	low	no	no	32	Czochr.
Piezoceramics		6…15	low	no	yes		sintering
Quartz	400	2.3	high	yes	no	32	hydroth.
$LiNbO_3$	300	8	low	no	yes	3m	Czochr.

Table 1: Physical properties of a selection of high temperature piezoelectric materials. The maximal operating temperature T max is given as an indication, as it actually depends on the application. Data from [1], and references therein.

5. Experimental

The piezoelectric properties of the crystals have been measured between room temperature and 800°C on small crystal plates using the direct piezoelectric effect by applying a dynamic force on the plate and by measuring the generated charge with a charge amplifier. This measurement was performed with several crystal cuts, hence allowing the complete characterization of the piezoelectric tensor d_{ij} [4]. The resistivity was measured on the same crystal plates using a DC electrometer. To address the stability issue, the resistivity measurement was repeated in vacuum (in-situ measurement). In addition, pressure sensors and accelerometers were built to test the properties of the sensors after ageing the sensors in a furnace at different temperatures (between 500 and 700°C) for up to 2000 hours (Fig. 2). The sensors were calibrated after each ageing stage.

The pressure sensors were built with a KI100 crystal element working in transverse mode (piezoelectric coefficient d'_{12}, rotated cut). Nickel based alloys were used to minimize creeping and the sensors were hermetically sealed. The signal was picked up with a mineral insulated cable. Three prototypes were built and intensively tested up to 600°C and aged over 2000 hours. The accelerometers were designed for temperatures up to 700°C. Several KI100 crystal rings were used in compression mode (piezoelectric coefficient d_{11}) and the sensor is ground insulated. In order to reduce electromagnetic interferences, the sensors were outfitted with a 2-pin connector with symmetrical capacitances pin to ground.

Fig. 2: M14 pressure sensor with integrated mineral insulated cable, Tmax = 600°C (left). Accelerometer, compressive design, with 2-pin connector, Tmax = 700°C (right).

6. Results and discussion

The sensitivity deviation of the PiezoStar® crystal KI100 is shown in Fig. 3. A special cut has been chosen to minimize the sensitivity deviation between room temperature and 800°C (± 4%). Also shown is the electrical resistance of a small plate like those which are used in the pressure sensors. At 800°C, the resistance only drops to 10^8 Ohm, which is a very high value compared to other crystals.

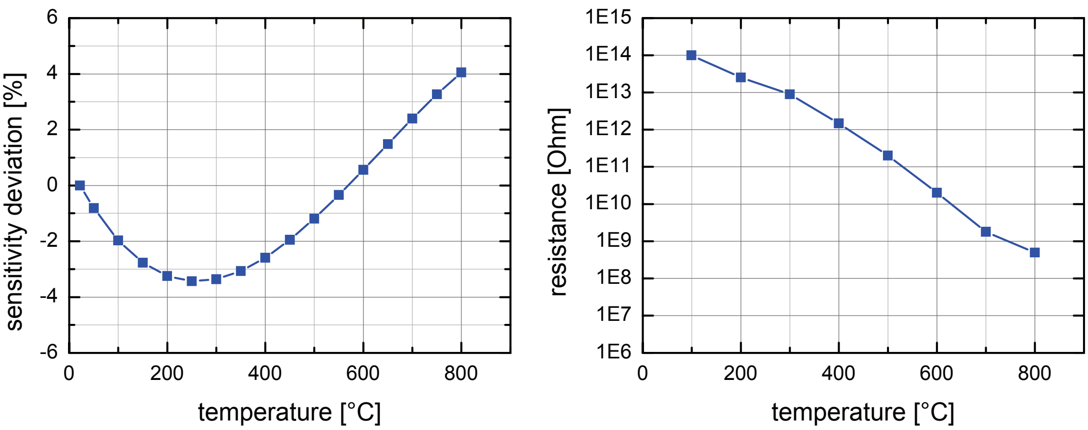

Fig. 3: Piezoelectric transverse sensitivity deviation (left) and electrical resistance in air (right) of the PiezoStar® crystal KI100 between room temperature and 800°C. The plate dimensions are 4.5 x 3 x 1.3 mm. Special rotated cut for minimal sensitivity deviation, sensitivity at room temperature: 5.3 pC/N.

The resistivity in vacuum ($p < 10^{-5}$ mbar) is shown in Fig. 4 (left). Above 400°C, the resistivity of KI100 is even higher than that of quartz. For comparison, the resistivity of KI85 and langatate has also been measured and plotted; the values are about 2 to 3 order of magnitude lower compared to KI100. The data shown in Fig. 4 (left) were taken upon heating. These are short time measurements (typically a few minutes) and they do not give any information about long-term stability. This is the reason why resistivity measurements were performed at 500°C, respectively 600°C, in vacuum for about 200 hours (Fig. 4, right). KI100 and quartz have stable properties at 600°C, whereas the resistivity of KI85 and langatate starts to decrease after a few hours at 500°C, eventually sinking to 10^5 Ohm after 200 hours. Clearly both KI85 and langatate can not be used for sensor applications at 500°C or higher, as their insulation properties are not stable enough. KI100 however has very good properties (the resistivity is high and stable) and is certainly one of the best piezoelectric materials for very high temperature applications.

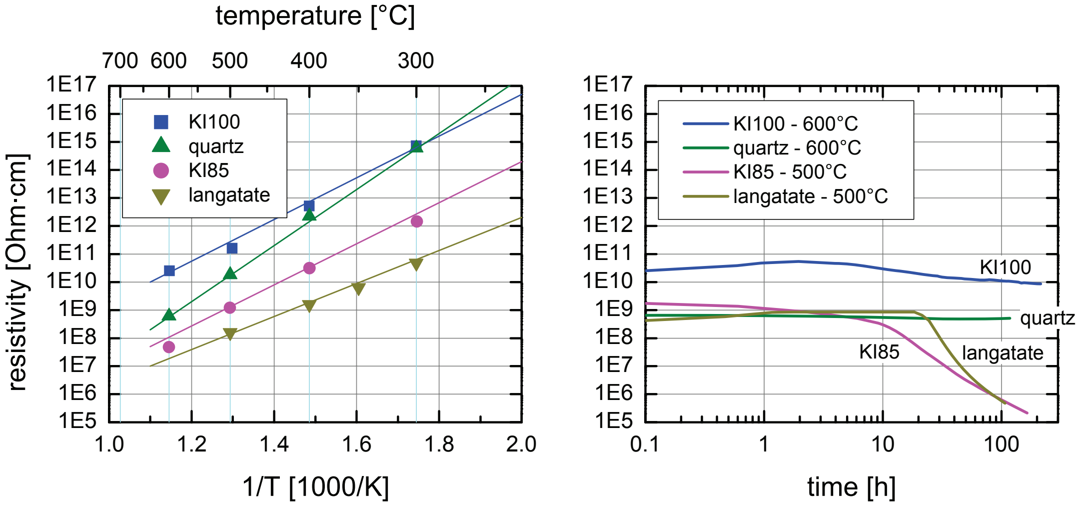

Fig. 4: Resisitivity as a function of temperature (left) and time (right) for the PiezoStar® crystals KI100 and KI85, quartz and langatate. The measurements were performed in vacuum (p < 10^{-5} mbar) on small discs (diameter 4.5 to 9.5 mm, thickness 0.5 to 1 mm).

The excellent properties of KI100 were confirmed with pressure sensors and accelerometers. Fig. 5 shows the sensitivity and resistance of the pressure sensors at different ageing stages. During the first 600 hours, the sensitivity decreases slightly after each ageing step. The overall sensitivity drop amounts less than 3% and is most probably due to sensor packaging setting rather than a degradation of the sensing element. Subsequent ageing steps did not have any detrimental impacts on the sensor properties; the resistance of the sensor remains very high during the whole experiment. Beside, the sensitivity lies within a very small band (-2 to +4%) over the whole temperature range. Note that these sensor prototypes were specifically designed for stability tests and this explains their relatively low sensitivity (45 pC/bar). The next sensor generation will have a sensitivity of about 250 pC/bar.

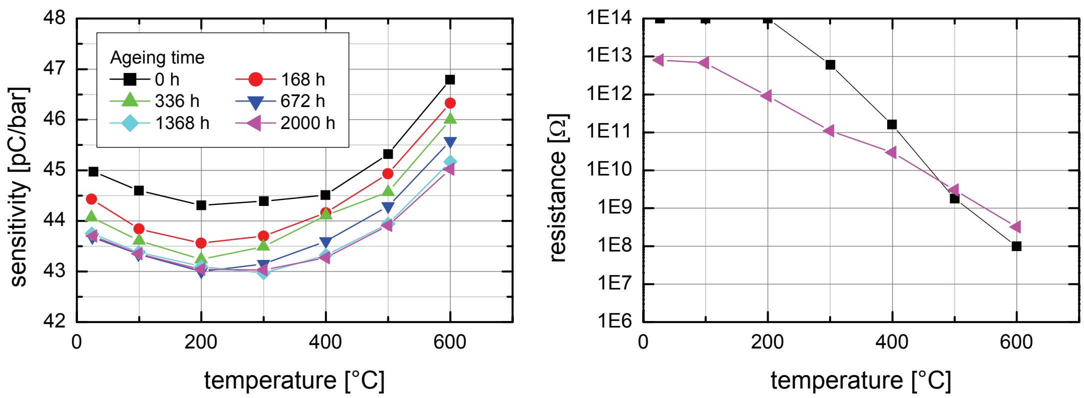

Fig. 5: Sensitivity and insulation resistance of the pressure sensor with KI100, measured after different ageing stages. Ageing was performed in a furnace at 600°C.

Similar stability tests were performed on the accelerometers. Ageing was performed first at 500°C for 336 h, then at 600°C for 672 h and finally at 700°C for 168 h. The calibration curves after each ageing stage are shown in Fig. 6. The resistance decreases during the first ageing steps, but then remains stable. The accelerometers have a lower insulation resistance than the pressure sensors. This is partly due to the geometrical factor of the sensing element, which is less favorable for crystal discs than for the transverse plates used in the pressure sensors.

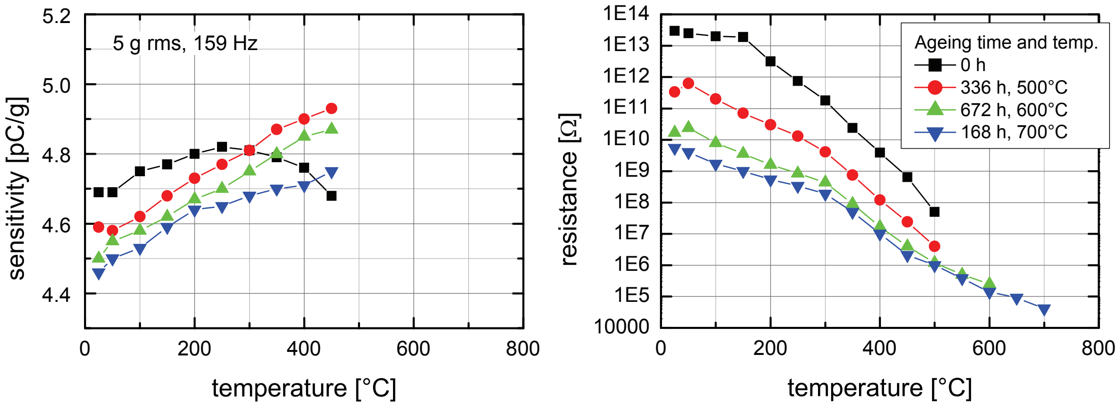

Fig. 6: Sensitivity and insulation resistance of the accelerometers with KI100, measured after different ageing stages. Ageing was performed in a furnace at 500, 600 and 700°C. The sensitivity was measured up to 450°C only.

7. Conclusion

High temperature stability tests (500°C to 700°C) have been performed on the PiezoStar® crystal KI100 to determine whether this new crystal is suitable as sensing element for pressure sensors or accelerometers. The results show that KI100 has outstanding insulation properties. The resistivity is very high and, more important, remains stable in reducing atmosphere up to at least 600°C, possibly 700°C. In addition, KI100 has a small temperature coefficient, it is not pyroelectric, and no twinning occurs up to the melting point, which makes KI100 one of the best piezoelectric material for high temperature applications. The feasibility of pressure and acceleration sensors for continuous operation at 600°C has been demonstrated. The potential to achieve even higher temperatures has not yet been fully exploited.

References

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