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Assessment of Material Properties of Gallium Orthophosphate Piezoelectric Elements for Development of Phased Array Probes for Continuous Operation at 580°C

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Abstract: In this paper, the thickness extension mode gallium orthophosphate single crystal elements were characterised using the impedance analyser. Impedance characteristics of piezoelectric elements were investigated at temperatures from 25°C up to 580°C at first and then at a constant temperature of 580°C for a period of 25 days. The resonant and anti-resonant frequencies extracted from the impedance characteristics, capacitance (measured at 1 kHz), density and dimensions of the gallium orthophosphate elements were used to calculate electromechanical, piezoelectric and elastic properties of these elements at high temperatures as a function of time. The tested gallium orthophosphate elements proved to possess very stable efficiency and sensing capability when subjected to high temperature. The results are very encouraging for proceeding with development of phased array probes using gallium orthophosphate, for inspection and condition monitoring of high temperature pipelines in power plants at a temperature up to 580°C.

1. Introduction

Today, a number of power plants (PP) are operating beyond their designed life. Rigorous standards, regulations and codes are in place by regulatory bodies to ensure structural health inspection is carried out periodically during planned outages. Since the inspections are performed during outages, the inspection techniques and technologies used are effective at room temperature. However, if a vital part of the PP needs to be monitored in service following an outage, such as a pipeline carrying superheated steam, it may present a problem, particularly for an aging PP. As an example, high temperatures (HT) and pressures experienced in these pipelines, can lead to creep, fatigue and corrosion type defects, which if undetected may have catastrophic consequences. For this reason, *in situ* condition monitoring techniques should be developed to retain reliability and extend the lifetime of aging PP.

Advanced ultrasonic techniques, such as Phased Array (PA) can be used to detect defects at room temperature. However, at HT operating conditions this technique cannot be applied due to lack of availability of HT probes for operation at temperatures up to 580°C in electrical PP [1]. The key challenge is to develop HT PA probes for continuous monitoring of defect growth over time, so that when the defect reaches a critical size the PP can be shut down and maintenance can take place before failure.

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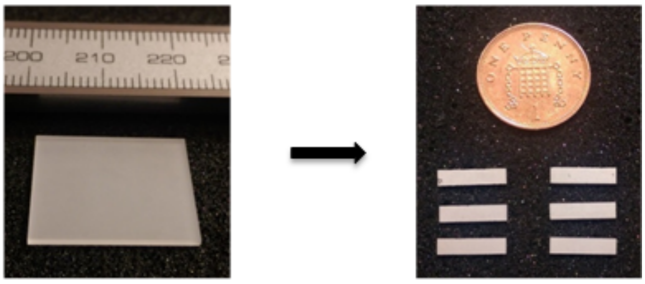
These PA probes use piezoelectric elements for generation and reception of ultrasound. Lead Zirconate Titanate (PZT) is the most commonly used piezoelectric for PA probes, but has a maximum operating temperature of around 180°C (half of the Curie temperature for PZT-5A) [2] which is not suitable for this application. Therefore, alternative piezoelectric materials need to be considered. A number of piezoelectric materials that possess high operating temperature have been reported [3], [4]. Piezoelectric nonferroelectric single crystals such as quartz (α-SiO_2), gallium orthophosphate ($GaPO_4$), langasite ($La_3Ga_5SiO_{14}$, LGS) and aluminium nitride (AlN) stand out because they exhibit no Curie temperature and no domain-related aging behaviour while showing good sensitivity and the ability to function over a broad temperature range up to very HT e.g. 800°C for the langasite crystals [4].

Due to excellent thermal stability of most of its material properties up to 970°C (>>580°C) as well as commercial availability of high quality high precision piezoelectric elements, single crystal $GaPO_4$ has been selected for further study [5].

Ideally the PA probes should function continuously in between planned outages. However, material properties of $GaPO_4$ for continuous operation at 580°C were not available in literature. Several methods, such as impedance method, the quasi-static method and the laser interferometry method are used to measure the material properties of piezoelectrics and they all provide high accuracy results [6], [7]. However, due to a fact that an impedance analyser was available for impedance measurements to be carried out on the $GaPO_4$ elements, it was decided to proceed with the impedance method. This method was used to determine the material properties of $GaPO_4$ single crystal elements at HT such as thickness coupling factor, charge coefficient and compliance and stiffness coefficients; these properties were derived from measured values of the resonant (f_r) and anti-resonant (f_a) frequency pair, capacitance (measured at 1 kHz), density and the dimensions of appropriately cut and excited $GaPO_4$ elements as outlined in the European Standard on Piezoelectricity [8]. From the derived material properties it is possible to draw conclusions about the efficiency, sensing capability and mechanical stability of the $GaPO_4$ elements at HT and consequently about the HT operation of the PA probe that would use these elements for the generation and reception of ultrasound.

2. Development of PA Elements

The $GaPO_4$ elements had to be manufactured before the measurements were carried out. The PA probe will use angled compression waves for the inspection of HT pipes. The way in which the piezoelectric element vibrates when stimulated by an electrical pulse depends upon the "cut" in the case of piezoelectric single crystals such as $GaPO_4$. The X-cut (equiv. to thickness extension mode for piezoelectric ceramics) of single crystals can generate and detect compression waves [9]; for this reason raw X-cut $GaPO_4$ plates were purchased from the manufacturer "Piezocryst Advanced Sensorics GmbH", Austria, to be appropriately shaped and coated with electrode material, as shown in Figures 1 a) and b). To build an efficient linear array of piezoelectric elements for the PA probe, the element dimensions must comply with the following: one dimension along the element and perpendicular to the thickness should be narrow for better directivity of the produced ultrasonic beam. The other dimension perpendicular to the above two directions should be large in order to increase the total element radiating area for meeting the power output requirement [10]. $GaPO_4$ elements with dimensions of 3 mm x 12 mm x 1 mm (width, length and thickness) were manufactured to meet those requirements, but at the same time to provide piezoelectric elements robust enough to manipulate with when performing impedance measurements at HT. The dimensions of the $GaPO_4$ elements that will be used in the HT PA probes will be different from the above dimensions, taking into account specification of the defects that want to be measured with the PA probes. Finally, the $GaPO_4$ elements were coated in the standard parallel electrode configuration, with a platinum layer of 100 nm considering the platinum thin film electrodes are used for HT up to 650°C due to their excellent electrical properties, high melting point and outstanding oxidation resistance [11].



a) **b)**

Figure 1: a) A raw $GaPO_4$ single crystal plate. **b)** The raw $GaPO_4$ plate was shaped into thin, slender rectangular bars to build an efficient linear array of piezoelectric elements for the HT PA probe.

3. Material Properties

The material properties of piezoelectrics are anisotropic and exhibit various values in different directions, depending on the cut in terms of piezoelectric single crystals. According to the step-by-step procedure for calculating a complete set of material properties for piezoelectric materials found in the European Standard on Piezoelectricity [8], for the X-cut single crystal elements vibrating in the thickness extension mode there are four relevant material properties that has to be derived: (i) thickness coupling factor k_t (electromechanical), (ii) charge coefficient d_{11} (piezoelectric) and (iii) compliance and stiffness coefficients, s^E_{11} and c^E_{11}, respectively (elastic). All of these four properties represent numerical measures of the efficiency, sensing capability and mechanical stability of the piezoelectric elements, as explained below.

3.1. Electromechanical Coupling Factor

The electromechanical coupling factor k_{ij} is a key parameter when describing the behaviour of piezoelectric materials, given that it expresses a numerical measure of efficiency for electro-mechanical conversion of the piezoelectric materials [12]. The first subscript to k denotes the direction along which the electrodes are applied; the second denotes the direction along which the mechanical energy is applied, or developed. The dimensions of a piezoelectric element dictate unique expressions of this factor. For piezoelectric elements such as the X-cut $GaPO_4$ elements shaped into thin, slender rectangular bars, whose surface dimensions are large relative to the thickness, the thickness coupling factor – k_t is the matching factor and it expresses the coupling between an electric field in direction 1 and mechanical vibrations in the same direction. Electromechanical coupling factors are found to range from 8% for quartz crystals to >70% for PZT ceramics [4].

3.2. Piezoelectric Charge Coefficient

The piezoelectric charge coefficient d_{ij} gives the ratio of the charge generated per unit of mechanical stress applied to a piezoelectric material, and conversely is the ratio of the mechanical strain developed by a piezoelectric material per unit of electric field applied. The first subscript to d indicates the direction of polarization generated in the material; the second subscript is the direction of the applied stress or the induced strain, respectively. The sensitivity needs to be sufficiently high so that the generated signal can be detected above the background noise. In practice, the generated signal is small and has to be enhanced by an appropriate charge or voltage amplifier [13]. The matching coefficient for the X-cut $GaPO_4$ elements vibrating in the thickness extension mode is the coefficient d_{11} where the subscript *11* denotes that both, the excitation of the element and the response of the element are in the same direction. Generally, typical values of the charge coefficient for piezoelectric materials range from about 2 pC/N for quartz crystals to about >700 pC/N for PZT ceramics [4].

3.3. Compliance and Stiffness Coefficients

The elastic compliance and stiffness coefficients, s^k_{ij} and c^k_{ij}, respectively relate the applied stress with the relative deformation of the material – the strain. These two coefficients are numerical measures for the mechanical stability of piezoelectric materials and ideally should not change with temperature. The matching compliance and stiffness coefficients of the X-cut $GaPO_4$ elements vibrating in the thickness extension mode are s^E_{11} and c^E_{11} coefficients, respectively, where the superscript *E* denotes a constant electric field, and the subscript *11* indicates that the direction of strain and the direction of stress are the same. For the *11* direction of piezoelectric materials, the *c* coefficient is the Young's modulus of elasticity.

3.4. Calculation of Material Properties from Measured f_r and f_a Values

Applying suitable modification, the mathematical relations from formulas presented within the European Standard on Piezoelectricity [8] were derived, considering the direction of positive polarization for piezoelectric ceramics vibrating in the thickness extension mode usually is made to coincide with the direction 3 (Z) of a rectangular system of 1 (X), 2 (Y) and 3 (Z) axes and for the piezoelectric single crystals vibrating in the same mode the direction of positive polarization will be the direction 1 (direction X / X-cut of a crystal).

The thickness coupling factor k_t can be derived based on the measured resonance and anti-resonance frequencies of piezoelectric elements, Equation 1:

$$k_t^2 = \frac{\pi}{2}\frac{f_r}{f_a}\cot\left(\frac{\pi}{2}\frac{f_r}{f_a}\right) \tag{1}$$

The piezoelectric charge coefficient d_{11} can be derived from the relation of the thickness coupling factor k_t, the dielectric coefficient ε^T_{11} and the compliance coefficient s^E_{11}, Equation 2:

$$d_{11} = k_t(\varepsilon^T_{11}\, s^E_{11})^{1/2} \tag{2}$$

The dielectric coefficient ε^T_{11} can be calculated from the "free" capacitance C^T measured well below the lowest resonant frequency e.g. at 1 kHz (*T* denotes the constant mechanical stress in the element), the thickness *t* of the piezoelectric element and the electrode area *A*, Equation 3:

$$\varepsilon^T_{11} = C^T\frac{t}{A} \tag{3}$$

The compliance coefficient s^E_{11} can be derived from the thickness coupling factor k_t and the compliance coefficient s^D_{11} where *D* denotes the constant displacement in the element, Equation 4:

$$s^E_{11} = \frac{s^D_{11}}{1-k_t^2} \tag{4}$$

The compliance coefficient s^D_{11} can be calculated directly from the measured anti-resonance value f_a, the density ρ (taken from the datasheet) and the length l (CTE taken into account) of the piezoelectric element, Equation 5:

$$s^D_{11} = \frac{1}{4\rho f_a^2 l^2} \tag{5}$$

Finally, the compliance coefficient c^E_{11} can be derived from the k_t factor and the compliance coefficient c^D_{11}, Equation 6:

$$c_{11}^{E} = c_{11}^{D}(1 - k_t^2) \tag{6}$$

And to calculate c^{D}_{11} one has to measure f_a, ρ and t, Equation 7:

$$c_{11}^{D} = 4\rho f_a^2 t^2 \tag{7}$$

4. Experimental Setup and Procedure

Five $GaPO_4$ elements were used for impedance measurements at HT. The thickness extension mode element has a compression velocity of 4356 m/s and calculated resonant frequency of 2.17 MHz. The dimensions at 25°C were determined using a digital caliper to an accuracy of 0.01 mm, and for HT the dimensions were calculated using temperature dependent coefficients of thermal expansion (CTEs) provided in the material datasheet. Density of the elements was also taken from the datasheet [14].
At room temperature, devices such as “smart tweezers” are usually used for the impedance measurements of piezoelectric elements [15]. However, those commercially available devices are not suitable for measurements at HT. Hence, a special holder for the $GaPO_4$ elements was designed to ensure that all the elements were tested in the same manner. The holder allowed electrical connection between an Agilent 4294A impedance analyser and the $GaPO_4$ elements under test.

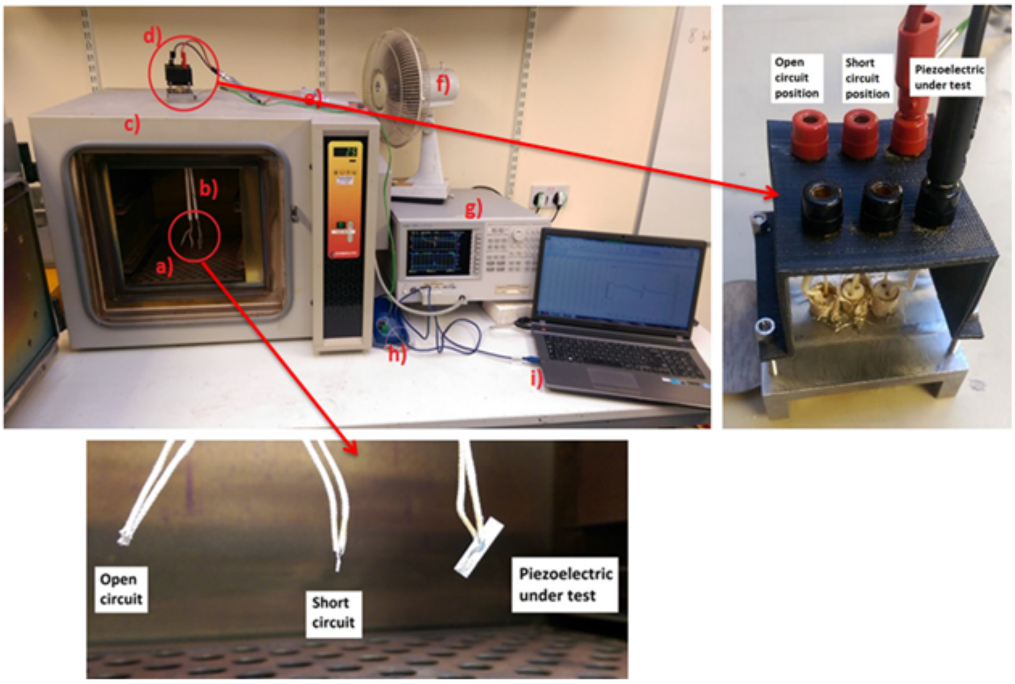


Figure 2: Experimental setup used for the impedance measurements of $GaPO_4$ elements at HT consisted of the following elements: a) piezoelectric element under test affixed to nickel conductors, b) alumina insulation tubing, c) Carbolite LHT6-30 oven, d) plastic panel with banana plug sockets for open and short circuit compensation, e) BNC test leads with Agilent 16048A test fixture, f) fen used for cooling of the outer part of the piezoelectric element holder, g) Agilent 4294A impedance analyser, h) temperature logger with K type thermocouple and i) computer to collect the data.

To establish a baseline, impedance characteristics of the $GaPO_4$ elements were recorded first at 25°C. Then, measurements were taken at 200°C, 400°C and 580°C. Finally, the temperature was kept at a constant value of 580°C for a period of 25 days in order to examine the material properties of the

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$GaPO_4$ elements at HT over time. Prior to proceeding with measurements at each temperature, compensation was carried out to remove the effect of parasitic impedance of the holder [16]. The f_r and f_a frequencies taken from the impedance characteristics, capacitance (measured at 1 kHz), density and dimensions of the $GaPO_4$ elements were used to derive material properties of the elements at HT.

5. Results and Discussion

5.1. High Temperature Material Properties

The material properties k_t, $d_{11,}$ s^E_{11} and c^E_{11} (averaged for five $GaPO_4$ elements) were derived and used to assess the effect of increasing temperature on the HT operation of $GaPO_4$ piezoelectric elements, Figure 3 and 4:

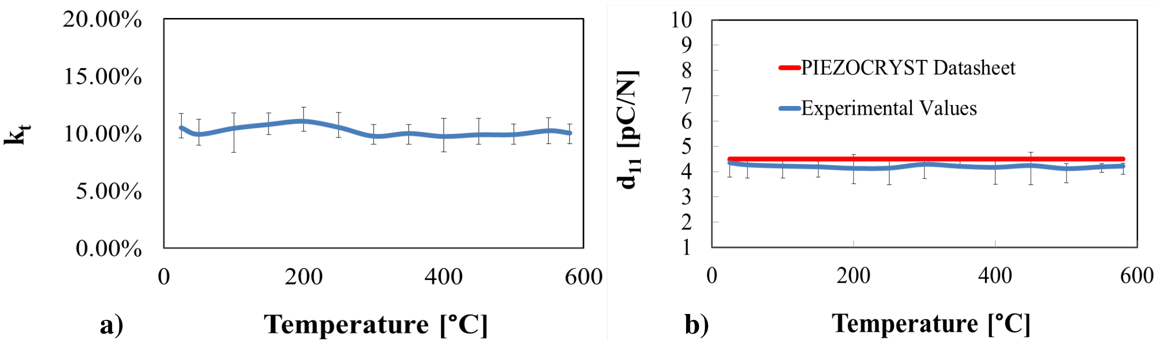

Figure 3: a) Thickness coupling factor k_t and **b)** Piezoelectric charge coefficient d_{11}, averaged for five $GaPO_4$ elements, as a function of temperature from 25°C to 580°C.

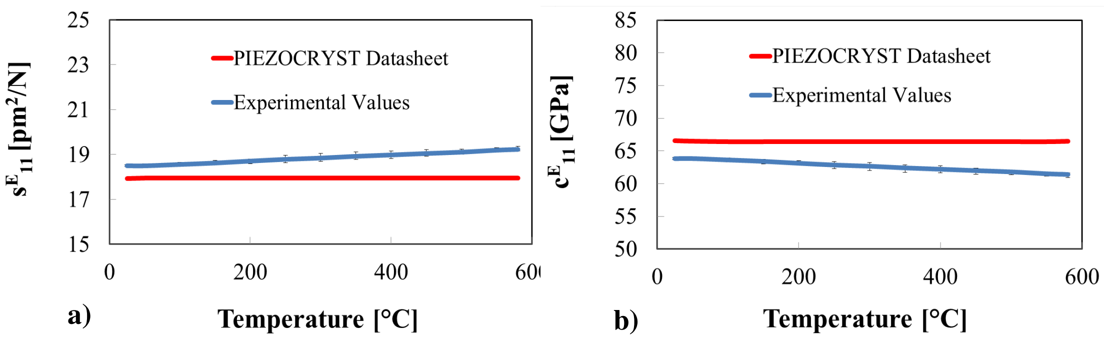

Figure 4: a) Elastic compliance s^E_{11} and **b)** Stiffness c^E_{11} coefficients, averaged for five $GaPO_4$ elements, as a function of temperature from 25°C to 580°C.

From Figure 3 a) it can be seen that the value of k_t remained almost constant at 10% from 25°C up to 580°C. This means that in this temperature range, the tested $GaPO_4$ elements can convert 10% of the energy delivered. In order to design a PA probe with broader bandwidth for better resolution, it is important for each of the $GaPO_4$ element to have high electromechanical coupling coefficient [10]. When comparing $GaPO_4$ elements to PZT ones which display conversion of up to 70% [2], the conversion of 10% seems to be very poor. However, it is important to keep in mind that e.g. PZT-5A elements cannot be used above around 180°C, where $GaPO_4$ elements were tested up to 580°C. From Figure 3 b) it can be seen that the derived d_{11} coefficient stayed constant at 4.2 pC/N in the whole tested temperature range. Same as with the k_t factor, the value of $d_{11} = 4.2$ pC/N when compared with PZT-5A which shows $d_{11} \approx 370$ pC/N is quite low; however the $GaPO_4$ elements showed very stable piezoelectric response far above the upper limitation temperature of PZT piezoelectrics. Finally, the calculated elastic properties proved very good mechanical stability of this piezoelectric material at up to 580°C, Figure 4 a) and b). The calculated s^{E}_{11} coefficient was 18.5 pm^2/N at 25°C and increased up to 19.22 at 580°C, the change was less than 3%. Furthermore, in the same temperature range, the calculated c^{E}_{11} coefficient deviated less than 4%, with the value of 63.84 GPa at 25°C and 61.42 GPa at 580°C. The comparison of the derived material properties with the datasheet values confirmed that the adopted measurement technique is reliable in determining the material properties of $GaPO_4$ elements at HT; thus, this technique can be used for the characterisation of $GaPO_4$ elements at HT in a long term.

5.2. *Long Term Material Properties at 580°C*

One $GaPO_4$ element was tested at 580°C for 25 days to obtain preliminary results on HT operation of $GaPO_4$ material in a long term in order to evaluate the feasibility of $GaPO_4$ elements for long-term continuous monitoring at HT. The results of the material properties at HT against time are shown in Figures 5 and 6:

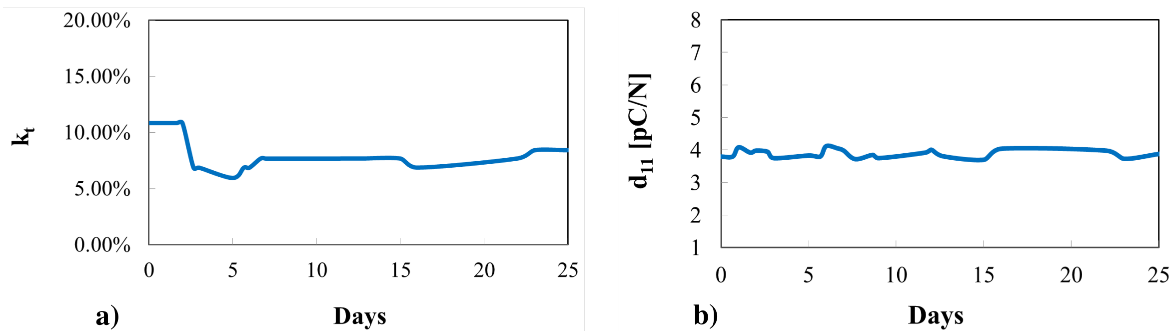

Figure 5: a) Thickness coupling factor k_t and **b)** Piezoelectric charge coefficient d_{11}, of a $GaPO_4$ element, as a function of time for 25 days at 580°C.

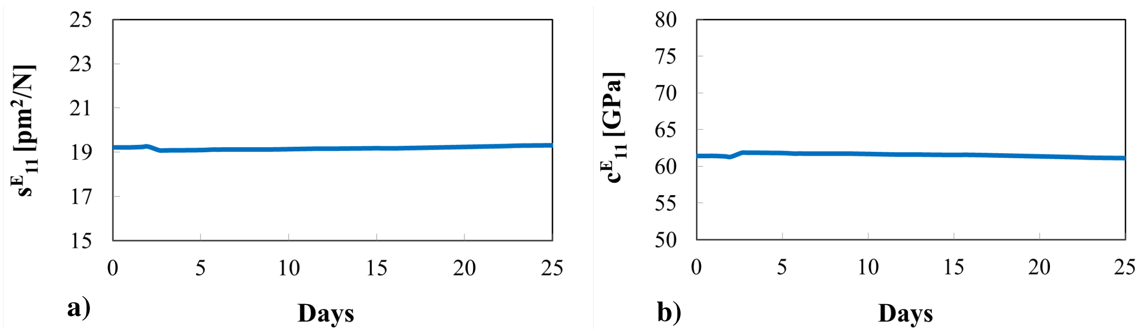

Figure 6: a) Elastic compliance s^{E}_{11} and **b)** Stiffness c^{E}_{11} coefficients, of a $GaPO_4$ element, as a function of time for 25 days at 580°C.

In Figure 5 a) it can be seen that the value of k_t factor decreased from the initial value of 10% in the first four days of testing and then stabilised at around 7.5% until the end of the test. According to literature, when compared to piezoelectric and elastic properties, the coupling factor is found to be insensitive to effect of temperature in single crystals [4]. This decline of 25% in the conversion of the energy delivered shows the opposite and more $GaPO_4$ elements need to be subjected to HT tests in a long term to validate the initial results. Figure 5 b) shows that the d^{E}_{11} coefficient remained at around 4 pC/N during the entire duration of the test confirming stable sensing capability of this piezoelectric material at HT. The derived elastic properties s^{E}_{11} of 19.2 pm^2/N and c^{E}_{11} of 61.6 GPa, stayed virtually unchanged at 580°C for 25 days, Figure 6 a) and b). After the 25-day period, the tested $GaPO_4$ element was cooled down to 25°C and impedance measurement was carried out again. The very pronounced f_r

and f_a peaks after the heat treatment confirmed that this piezoelectric material could potentially be used in development of PA probes for continuous operation at HT.

6. Conclusions

In this study the material properties have been derived for five $GaPO_4$ elements using the impedance method as outlined in the European Standard on piezoelectricity. In order to ensure that all the $GaPO_4$ elements were tested in the same manner, a special holder was designed. The five $GaPO_4$ elements proved to possess very stable material properties from 25°C up to 580 °C and one $GaPO_4$ element stayed very stable when subjected to 580°C for 25 days. However, the very low measured values of k_t factor of 7.5% (PZT $k_t \approx 70\%$) and d^E_{11} constant of 4 pC/N (PZT-5A $d^E_{11} \approx 370$ pC/N) urge caution and further study is needed. One of the proposed solutions to utilise this very low percentage of the converted energy (low efficiency) and low sensing capability when using $GaPO_4$ elements for generation and reception of ultrasound is development of appropriate low-noise signal amplifiers. Also, the measured electrical impedance of the $GaPO_4$ elements was in the order of kΩ which is several orders of magnitude higher than the load on the standard BNC cables (50Ω). The electrical impedance mismatching can affect the ultrasonic probe noise performance, driving response, bandwidth and sensitivity. Appropriate matching circuits need to be developed to overcome this issue.

7. Future Work

More $GaPO_4$ elements will be tested at HT for a period of 25 days in order to provide sufficient evidence to prove the reliability of such elements. Fabrication of $GaPO_4$ elements to achieve higher frequencies up to 5 MHz to be able to detect smaller defects should be possible and will form part of future effort. The next step in this study will be to simulate the ultrasonic propagation in order to assess the transmission and reception quality of the $GaPO_4$ array elements, both at room and HT operating conditions. Finally, PA probes will be built using $GaPO_4$ elements and subjected to laboratory and field trials.

8. Acknowledgements

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