ULTRASONIC TRANSDUCERS FOR HIGH TEMPERATURE APPLICATIONS IN ACCELERATOR DRIVEN REACTORS

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Abstract

The paper is devoted to description of developed various ultrasonic transducers suitable for long term measurements in a liquid Pb/Bi alloy, which is used as a coolant in accelerator driven systems.

The results of comparative experimental investigations of the developed transducers of different designs in a liquid Pb/Bi alloy up to 450°C are presented. The radiation robustness was assessed by exposing the transducers to high gamma dose rates in one of the irradiation facilities at SCK•CEN.

The experimental results proved that the developed transducers are suitable for long term operation in harsh conditions.

Introduction

In some nuclear reactors or accelerator driven systems (ADS) the core is intended to be cooled by means of a heavy liquid metal (HLM). In contrast to water-cooled reactors, it is not possible anymore to inspect optically inner reactor parts, which are submerged in the HLM. For safety and licensing reasons, a visualisation method, based on application of ultrasonic waves, has thus to be developed. The Belgian Nuclear Research Centre (SCK•CEN) is engaged in preparing the design of the ADS system MYRRHA and has started a co-operation with the Ultrasound Institute of Kaunas University (Lithuania) in order to develop an ultrasonic visualization system [1].

This system will exploit piezoelectric transducers submersed in the liquid lead-bismuth alloy. These transducers will be exposed to very harsh conditions. The temperature will be in the range 160 - 450 °C and the gamma dose rate could be up to 30 kGy/h. In addition, the transducer housing and cables will be subject to liquid metal corrosion. There are known various modifications of high temperature ultrasonic transducers, but they are not intended for such harsh conditions [2, 3, 4].

For the construction of the first prototype ultrasonic transducers, various technical obstacles had to be overcome: acoustic coupling between ultrasonic transducer and the liquid metal, acoustic coupling between the piezoelectric element and the sensor protector at all temperatures, leak-tight cable penetrations, etc. All these problems were investigated and solved. Prototypes with different high temperature piezoelectric materials were developed: PZT, bismuth titanate ($Bi_4Ti_3O_{12}$), lithium niobate ($LiNbO_3$), gallium orthophosphate ($GaPO_4$) and aluminum nitride (AlN). For acoustic coupling with the metal alloy, it was proposed to coat the active surface of the transducers by diamond like carbon (DLC). Such coatings could be also used to prevent a metal corrosion. The results of comparative experimental investigations of the developed transducers of different designs in a liquid Pb/Bi alloy proved that the transducers are suitable for long term operation in harsh conditions.

Statement of the problem

The ultrasonic transducers used for imaging of interior of the ADS system MYRRHA must meet the following requirements:

- Continuous operation at high temperatures (up to 450^{o}C);
- Corrosion-proof in aggressive liquid Pb/Bi alloy;
- Resistibility to radioactive (γ and neutron) radiation;
- Good performance in a pulse mode.

Problems to be solved are the following:

1. Selection of piezoelectric materials suitable for operation at elevated temperatures and under strong radioactive irradiation.
2. Acoustic coupling of a piezoelectric element to protector and backing at high temperatures.
3. Durable and stable acoustic coupling of an ultrasonic transducer to the liquid metal (Pb/Bi) alloy.

In order to provide a durable acoustic coupling between the piezoelectric element, protector and backing it is necessary to find out a suitable bonding or coupling technologies.

Piezoelectric elements

Piezoelectric materials used in high temperature ultrasonic transducers must possess the Curie temperature higher than 450°C. It is necessary to keep in mind that a recommended highest operational temperature is lower than the Curie or phase transition temperature. That significantly restricts the number of possible materials. The high temperature materials selected for investigation are listed in Table 1. Some materials, such as lead zirconate titanate (PZT) and lithium niobate ($LiNbO_3$), are well known, but performance of bismuth titanate ($Bi_4Ti_3O_{12}$), gallium orthophosphate ($GaPO_4$) and aluminum nitride (AlN) piezoelectric elements in above-mentioned harsh conditions was not yet determined.

Table 1: Piezoelectric materials for bulk wave high temperature ultrasonic transducers

Piezoelectric material	Type, manufac-turer	Curie tempe-rature, T_C,°C	Electro-mechanical coupling coefficient
PZT	Pz23, Pz27 Ferroperm	350	0.45-0.47
$Bi_4Ti_3O_{12}$	Pz46 Ferroperm	650	0.20
$GaPO_4$	AVL LIST GmbH	970*	0.15
$LiNbO_3$	36° Y- cut	1210	0.49
AlN	University Dayton	>2200#	0.2

*phase transition temperature
melting temperature

Design of the transducers

High frequency piezoelectric elements (f_0=5MHz) are rather thin, therefore they could not stand alone against an external pressure created by the liquid metal alloy. Especially very thin are AlN films (f_0>(15-25)MHz); therefore they are deposited on a supporting substrate. In order to overcome this problem this substrate is exploited as a backing and a damping body, enabling to get a wide bandwidth and short pulse duration without multiple reflections inside backing. In order to achieve this goal, as the backing a metallic cylinder with a concaved back surface is used (Figure 1).

In order to protect a thin piezoelectric element from the aggressive alloy and external pressure a thin (λ/4 or λ/2) metallic membrane (Figure 1) or a metallic waveguide were used. The performance of these two different designs is very different, especially from the point of a wiev of pulse and frequency responses.

Therefore, experimental investigations were carried out with the both types of transducers.

Acoustic coupling between the piezoelectric element, metallic protector and backing in the whole temperature range was achieved using or high melting temperature solder (up to 310°C), or a dry acoustic contact (up to 450°C). In the last case in order to get a good coupling, the contacting surfaces were polished up to optical quality, which was checked by Newton's ring method. After that all elements of the transducer were pressed together. Quality of the acoustic contact may be improved significantly inserting thin high purity (99.99%) golden films between piezoelectric element, protector and backing. For comparison some experiments were carried out using as a couplant high temperature silicon oil.

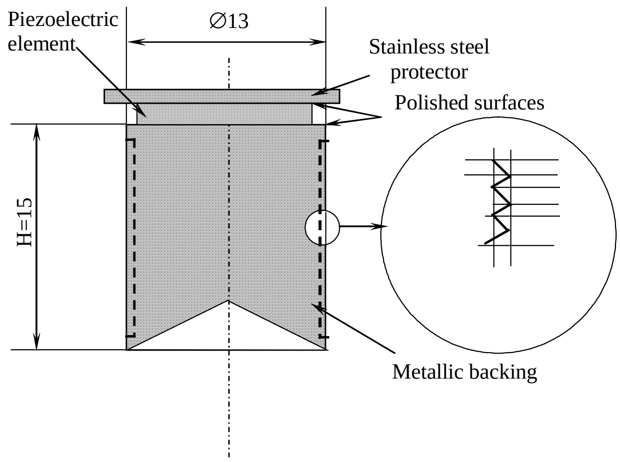

Figure 1: Monolithic transducer design with a metallic backing

Experimental results

Results of comparative experimental investigations of ultrasonic transducers with a thin metallic protector and waveguide are given in Table 2. In the case of a thin protector experiments were performed in water in a pulse-echo mode measuring the reflected ultrasonic pulses. Efficiency of the transducers was estimated by the transfer coefficient:

$$K = \frac{U_{r\max}}{U_{IN\max}},$$

where $U_{IN\max}$ is peak-to-peak amplitude of the excitation pulse, $U_{r\max}$ is the amplitude of the ultrasonic pulse reflected by a flat titanium reflector placed in water at 45mm from the transducer. Note, that in the liquid metal alloy the efficiency of the transducers was higher, because the acoustic impedance of the alloy is higher than that of water. In 50 mm steel waveguides for measurements the pulses reflected by a free end of the waveguide were used.

Resonance frequency of all piezoelectric elements was 5MHz, except the case of aluminium nitride.

Transducers were soldered or pressed to the thin steel protector using silicone oil. The transducers with a dry acoustic contact possess a similar transfer coefficient.

Table2: Efficiency of different ultrasonic transducers in a pulse-echo mode without electrical matching

Piezoelectric material	f_0, MHz	Transfer coefficient K, 10^{-4}	
		With a thin protector in water	In steel waveguide
Pz27	5	33	840
Pz46	5	13	37
$LiNbO_3$	5	-	60**
$GaPO_4$	5	-	<1**
AlN	15	0.14*	-

* direct contact

** with electrical matching K=211·10^{-4} ($LiNbO_3$) and K=27·10^{-4} ($GaPO_4$)

High temperature experiments

The developed transducers were investigated in the liquid Pb/Bi alloy at various temperatures. During the experiments, ultrasonic pulses reflected from a planar titanium reflector placed in the liquid Pb/Bi alloy were recorded. The titanium reflector was chosen because titanium is not wetted by the alloy what enabled to get a reflection coefficient close to 1.

Experimental results for the bismuth titanate Pz46 transducer are presented in Figure 2. The piezoelectric element Pz46 was soldered with a high temperature solder both to the stainless steel (AISI-316) polished $\lambda/2$ protector and to the metallic backing.

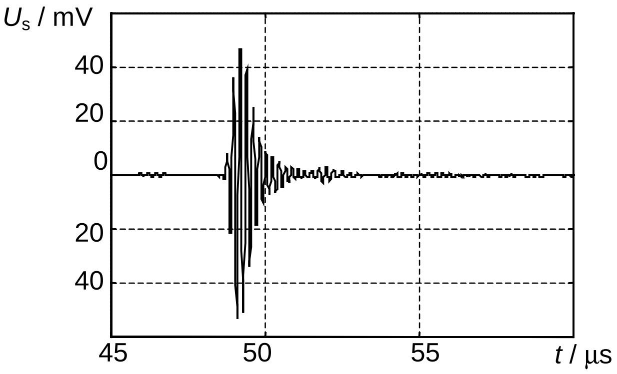

Figure 2: Pulse response of bismuth titanate Pz46 ultrasonic transducer with a thin ($\lambda/2$) protector in the Pb/Bi alloy at 160ºC

In the case of waveguide type transducers two different measurements for each transducer were performed. During the first measurement, the pulses reflected by the free end of the waveguide in air were recorded. During the second measurement the transducer was immersed into a liquid metal alloy and the pulse reflected by a planar titanium reflector placed at 10 mm from the tip of the waveguide was recorded.

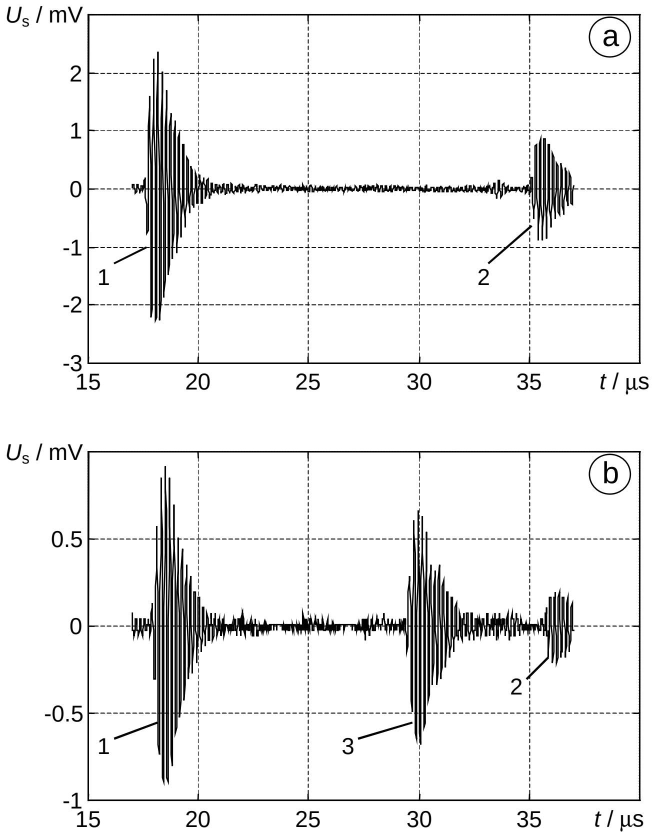

Figure 3: Ultrasonic waveforms of the waveguide type transducer with the $GaPO_4$ crystal: a - the first (l) and the second (2) reflections in the waveguide before immersion in the liquid Pb/Bi, (in air at 20°C); b – after the immersion into the liquid Pb/Bi at the temperature 250°C

Experimental results obtained with the 5MHz $GaPO_4$ type transducer are presented in Figure 3. The $GaPO_4$ crystal was coupled to the AISI 316 waveguide. After immersion into the liquid Pb/Bi the signal 3 (Figure 3b) appears, which is the reflection from the titanium reflector.

The results presented clearly indicated that both types of the transducers may be used in a high temperature environment, but bismuth titanate Pz46 transducers with a thin protector possess a higher efficiency.

A very serious problem is acoustic coupling between the front surface of the transducer and liquid metal alloy, which under usual conditions is not wetting a metallic protecting membrane. This problem was solved by polishing and (or) coating front surface of the transducer by diamond like carbon (DLC) film. That enabled to get a stable long-lasting (>1000 hours) acoustic contact between the transducer and the liquid Pb/Bi alloy when influence of the Pb/Bi oxides was prevented.

Quality of the acoustic contact was evaluated using the transducer with a waveguide. The results obtained during the first 150 hours are presented in Figure 4.

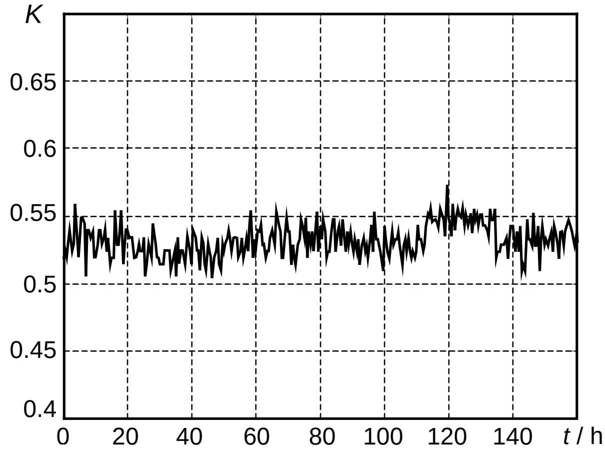

Figure 4: Time variation of the acoustic contact with the liquid Pb/Bi alloy at the temperature 200°C.

The quality of the acoustic contact was estimated by the ratio $K=U_2/U_1$, where U_1 is the amplitude of the signal reflected by the waveguide end, U_2 is the amplitude of the signal reflected by the titanium plate.

Irradiation results

The irradiation was carried out in the underwater gamma irradiation facility BRIGITTE at SCK•CEN in Mol in channel A2. Irradiation was performed by radioactive isotope ^{60}Co. The total dose received by the transducers was up to 22.7 MGy.

During experiments the transfer coefficient was measured. The results of experiments are summarized in Table3.

Table3: Resistance to γ radiation

No.	Transducer type	Total γ dose, MGy	Resistibility
1	Pz27	20	Good
2	Pz46	22.7	Good (Saturated reduction 4%)
3	$LiNbO_3$	20	Good
4	$GaPO_4$	22.7	Reasonable (Unsaturated reduction 13%)
5.	AlN	18.7	Very good (unaffected)

The bismuth titanate transducer experienced only a modest, saturated (4 %) decrease of the transfer coefficient, while the efficiency of the $GaPO_4$ transducer decreased by 13 %, without a clear indication of saturation.

The PZT, bismuth titanate and lithium niobate transducers showed no noticeable changes of pulse responses and transfer coefficients.

Conclusions

The investigation carried out showed that there are a few piezoelectric materials which are suitable for a long term operation in the liquid Pb/Bi alloy without cooling. At the moment the best material from the point of view of thermal and radiation robustness is the bismuth titanate Pz46. Other novel materials like aluminum nitride or gallium orthophosphate demonstrate good thermal stability ($GaPO_4$) or gamma radiation robustness (AlN), however their electroacoustic efficiency is significantly lower.

Other two serious problems are acoustic coupling in a wide temperature range between a piezoelectric element, protector and backing and a long-term stable acoustic contact between a front surface of an ultrasonic transducer and the liquid Pb/Bi alloy. For both of these problems solutions were proposed.

The investigations of the developed transducers of different designs in a liquid Pb/Bi alloy proved that they are suitable for long term operation in corrosive environment at elevated temperatures.

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References

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