International Journal of Scientific and Technical Research in Engineering (IJSTRE)
www.ijstre.com Volume 7 Issue 3 ‖ May-June 2022.
ISSN: 2581-9941

Investigation of ultrasound assisted soldering of SiC ceramic by Zn-Al-Ga high-temperature solder

Igor Kostolny[1], Roman Kolenak[1], Alexej Pluhar[1], Tomas Melus[1], Paulina Babincova[1] and Matej Pasak[1]

[1]*(Slovak University of Technology in Bratislava, Faculty of Materials Science and Technology in Trnava, Jana Bottu No. 2781/25, 917 24 Trnava, Slovak Republic)*

Abstract: *The work is oriented to research of ultrasonic soldering of SiC ceramics by use of Zn5Al1Ga solder. The solder type Zn5Al1Ga was studied by DSC analysis, while two endothermic reactions were observed. The first took place at the temperature of 243.3 °C and the second at the temperature of 370.9 °C. Complete melting of solder has occurred at the temperature of 395.5 °C. The tensile strength of Zn5Al1Ga solder has attained the value of 83 MPa. The XRD analysis has revealed the* $Al_{0.95}Ga_{0.05}$ *compound in the solder and the intermetallic phase of* $Al_{3,21}Si_{0.47}$ *in the boundary of SiC/Zn5Al1Ga joint. The shear strength of soldered joint of SiC/Zn5Al1Ga/SiC has attained the value of 54 MPa.*

Keywords - *microstructure soldering, SiC ceramic, ultrasonic soldering, microstructure.*

I. INTRODUCTION

The semiconductor parts destined for the printed circuit boards in electronics are nowadays joined mainly by use of Sn-based solders, which however exert very low melting point [1-3], namely the temperature lower than 250 °C. Regarding this fact, the tin solders are not capable to meet the requirements for modern equipments of power electronics servicing at elevated temperatures, what requires further development of soldering alloys applicable in this field, what thus creates the space for high-temperature soldering alloys. The Zn-Al based alloys exert excellent thermal and electrical conductivity and outstanding mechanical properties, what makes them suitable adepts for high-temperature soldering, applicable in a broad scope mainly in the electro-technical industry.

The research team from the Harbin Institute of Technology [4] has dealt with soldering SiC ceramics by use of ultrasound. Two types of solders were applied, namely type Zn5Al3Cu and Zn5Al. The effect of ultrasound on microstructure and mechanical properties of soldered joints was studied. The results have shown that ultrasound supports the wetting of parent metals and thus the bond is formed just in several seconds. In the case of Zn5Al3Cu solder the shear strength of soldered joints was approximately 139 MPa, due to occurrence of a great amount of η-Zn phases. With prolonging time of ultrasound acting the grain refining was observed. The shear strength of Zn5Al solder was approximately 102 MPa, however, with prolonging time of ultrasound acting the strength increase even up to the value of 138 MPa was observed, what corresponded to Zn5Al3Cu solder. Improvement of mechanical properties was closely related with a significant grain refining and the change in eutectic structure of materials joined.

Zhang et al. from the Wuhan University of Technology [5], have studied the soldered joints fabricated by the technology of ultrasonic soldering by use of Zn3Al solder. The Zr-based bulk metallic glass and the alloy type 1060 Al were used as substrates. They were able to fabricate the soldered joints free from cracks or voids. At ultrasound acting for 64 seconds a reaction layer was formed on the boundary of Zr-BMG/Zn3Al joint. Due to reduced concentration of Al in the solder/Zr-BMG boundary a composite layer of Zn50Al25Zr25 was formed. The results of shear strength measurement have shown that the joint fabricated by ultrasound acting for 64 seconds has attained the highest shear strength – namely 68 MPa.

Other works of authors [6-12] have dealt with the research of Zn – based solders for higher application temperatures. The subject of research were the solders with addition of other elements for improvement of their properties. Besides the change in melting point in dependence on the content of alloying elements also the change in mechanical properties of the studied alloys was observed. The addition of Mg and Ga to Zn-Al solder and study of its properties was dealt with by authors [13]. The solder was tested for die-attaching applications.

The melting point of solder was lowered to 347 ° C. Decreasing of micro-hardness with increasing temperature was observed. Authors [14] have studied the fluxless soldering. The addition of Ag to the structure of Zn-6Al solder resulted in formation of an intermetallic phase type $AgZn_3$. The melting point of solder was 380 ° C.

This work deals with the study of soldering SiC ceramics by use of Zn-Al-Ga based solder. The work is oriented to quality assessment of soldered joints fabricated by use of a selected solder and technology of ultrasonic soldering. The analysis is aimed to solder/substrate boundary and to strength measurement of fabricated joints. Also the fractured surface of joints after shear strength tests is assessed.

II. EXPERIMENTAL METHOD

The solder type Zn5Al, containing 5 wt. % Al in soldering alloy was prepared from pure Zn and Al elements. This soldering alloy is ranked among the eutectic solders. In its manufacture also a third element (Ga) was added in the amount of 1 wt. % to soldering alloy. The chemical composition of used solders is given in Table 1. Schematic representation of solder layout is shown in Fig. 1. The zinc alloys are typical with their high thermal conductivity and reliability depending on other alloying elements. This type of solder is characterized as the soldering alloy for higher application temperatures destined mainly for the electro-technical industry. Solder manufacture in as-cast condition was realized in a vacuum oven with resistance heating. The procedure was as follows: The working temperature used in manufacture was 900 °C. At holding this temperature for the time of 20 min, homogenization of soldering alloy took place. The alloy was subsequently slowly cooled down in vacuum oven at the cooling rate of 14 °C/min.

The following substrates were used in experiment:

- Ceramic SiC substrate in form of a square in dimensions 10 x 10 x 2 mm.
- Ceramic SiC substrate in form of a disk in dimensions Ø 15 x 2 mm.

Tab. 1 Chemical composition of solders

Solder designation	**Chemical composition of solder [wt %]**
Zn5Al	95 % Zn, 5 % Al
Zn5Al1Ga	94 % Zn, 5 % Al, 1% Ga

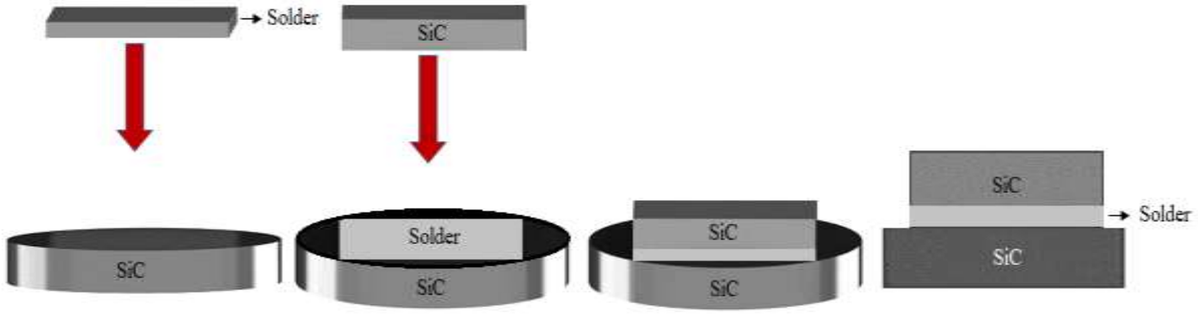

Fig.1 Schematic representation of layout of substrates and solder

Solder activation was realised via ultrasonic equipment type UT2 – ultrasound transducer making use of a piezoelectric system with a titanium sonotrode in diameter of Ø 3 mm. Ultrasound parameters are given in Table 2. The soldering temperature was by 20 °C higher than the temperature of molten solder. The soldering temperature was continually measured on the hot NiCr/NiSi plate by use of a thermocouple.

Table 2 Ultrasound parameters

Výkon ultrazvuku	[W]	400
Pracovná frekvencia	[kHz]	40
Amplitúda	[µm]	2
Spájkovacia teplota	[°C]	390
Čas pôsobenia ultrazvuku	[s]	5

The soldered joints are regularly subjected to thermo-mechanical loading due to temperature fluctuations and shear forces. The experimental joints were subjected to shear strength measurements on the equipment type LabTest 5.250SP1-VM and their values were mutually compared. The shear strength was measured by use of a special jig which schematic representation is shown in Fig.2. The measurement was realised up to total rupture of specimen.

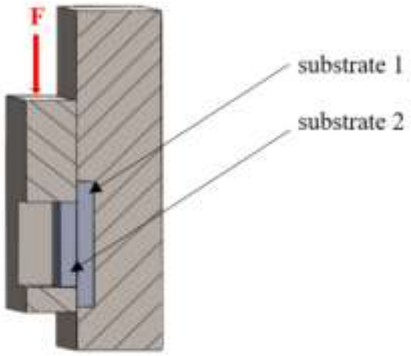

Fig.2 Schematic representation of a jig for shear strength measurement

The soldering process is realised on a hot plate at the presence of ultrasonic vibrations. The working procedure consisted in degreasing of SiC substrates, which are laid on the hot plate. Heating of the hot plate is set in accordance with the melting point of soldering alloy used. Subsequently a small amount of soldering alloy is deposited on the substrate surface. Next step consists in immersing the sonotrode of ultrasonic equipment into molten solder for the time of 5 s. The solder is thus activated due to action of ultrasonic vibrations resulting in disruption of oxides in the soldering alloy, which are gradually getting on the solder surface. The substrates with deposited solder, prepared in such a way, are then brought together and the bond is thus formed. These joints are then cooled down in the air at room temperature.

Metallographic preparation consisted in processes of grinding, polishing and etching of the embedded specimens. The specimens were inserted to a jig of equipment destined for grinding. Grinding was performed by use of SiC emery papers with the grain size of 600, 1200 and 2400. The grinding paper was during grinding supplied with water, ensuring thus the washing-off the debris at grinding. The grinding process with each paper lasted 3 minutes. After grinding, the polishing process followed by use of polishing disks with diamond emulsions with the particle size of 9 µm, 6 µm, 3 µm and 1 µm. It was performed for 4 minutes with each emulsion. After polishing the etching process followed. Etching was performed by use of etchant type NITAL with the chemical composition of 2 ml HNO_3 and 98 ml ethanol for the time of 4 s.

III. RESULTS

III.1 DCS Analysis

Fig. 3 shows the results of DSC analysis of Zn5Al1Ga solder (red colour). The onset of the first endothermic reaction at heating of Zn5Al1Ga solder was observed at the temperature of 243.3 °C. This temperature means the first eutectoid transformation. The second reaction has occurred at the temperature of 370.9 °C. This temperature (onset) characterized the melting of eutectic mixture of solid solutions (Zn)+(Al). The last peaks represent the energy necessary for melting of all phases contained in the solder. Complete melting of solder was achieved at the temperature of 395.5 °C (Zn5Al1Ga).

The soldering temperature is selected approximately at 20 °C above the melting point of soldering alloy. The soldering temperature selected for Zn5Al1Ga solder is 390 °C.

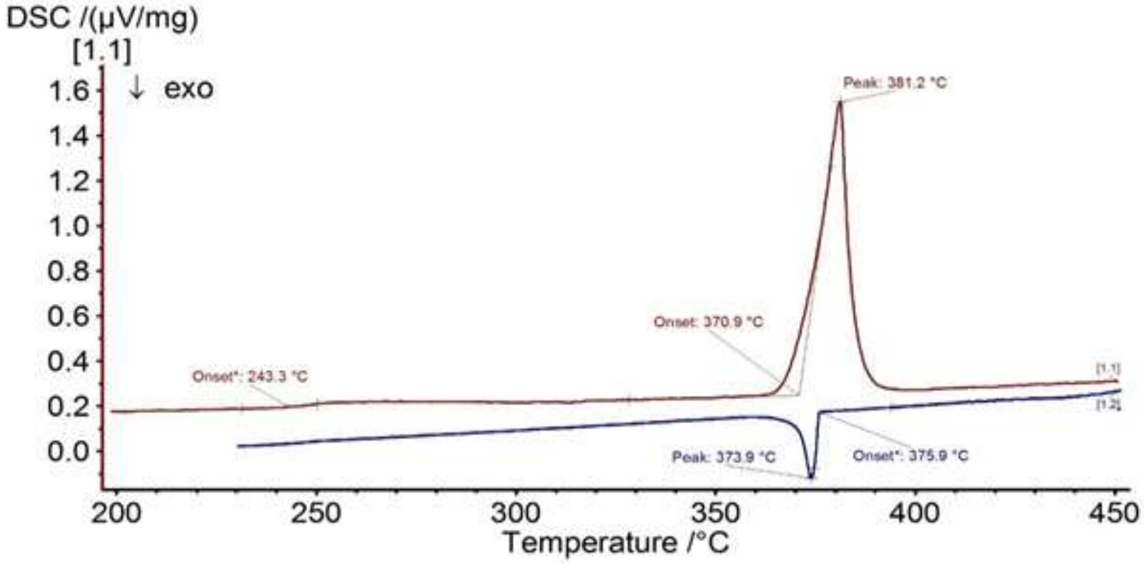

Fig.3 DSC analysis of Zn5Al1Ga solder

III.2 Strength measurement of solders

For assessment of a soldering alloy, the static tensile test is used. The average values from the measurement of individual solders is shown in the graph in Fig.4. For comparison of Zn5Al1Ga solder also the measurement on a binary solder type Zn5Al was performed, in order to enable the comparison of effect of added element on the resultant strength of the experimental alloy.

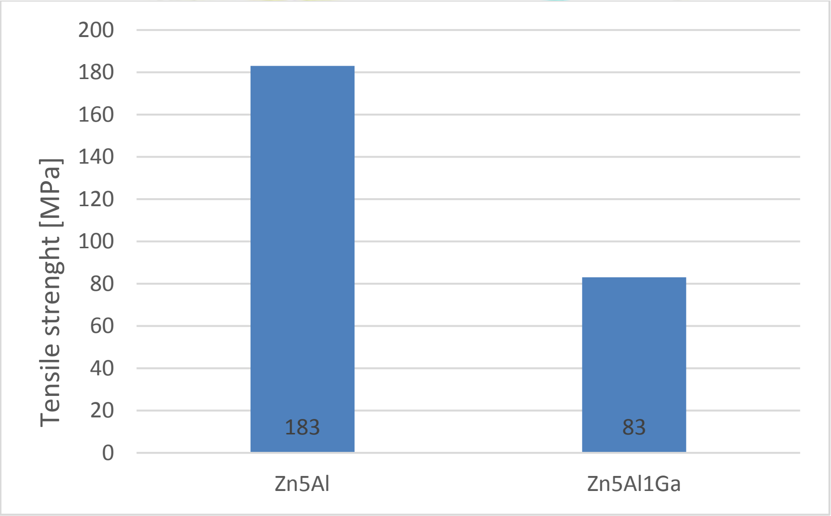

Fig.4 Tensile strength of experimental soldering alloys

The results clearly show that at addition of just 1 wt. % Ga to soldering alloy its strength was reduced. Whilst the gallium reduces the solder strength almost by half (45 %), it occurs in the solder just as the $Al_{0.95}Ga_{0.05}$ phase, eventually dissolved in Zn or Al. That´s why the gallium affects the strength of soldering alloy. The specimens for tensile test were of standardised shape and standardised dimensions, as shown in Fig. 5. All dimensions given in Fig.5 are in mm.

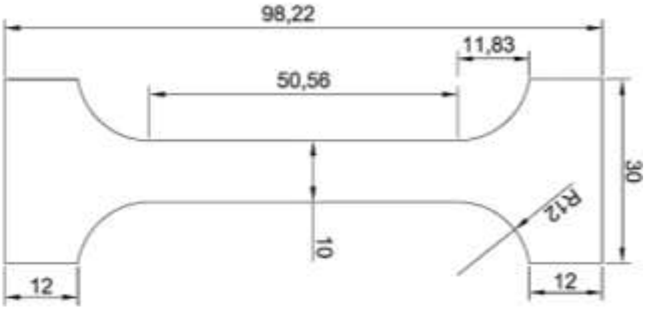

Fig. 5 Shape and dimensions of the test piece for tensile test

III.3 Solder microstucture

EDX analysis shows the presence of Ga (violet colour) in experimental solder, which is seen in the map of elements in Zn5Al1Ga alloy – Fig. 7. The microstructure of soldering alloy is shown in Fig.6. The solder preserved its basic structure of Zn-Al, which consists of a solid solution (Zn) and eutectics (Zn) + (Al). Distribution of gallium in the soldering alloy is uniform over entire volume of solder.

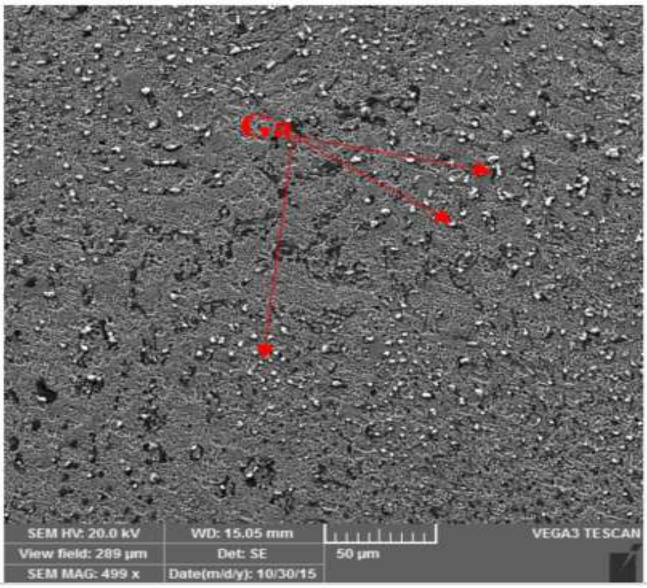

Fig.6 Microstructure of soldering alloy type Zn5Al1Ga

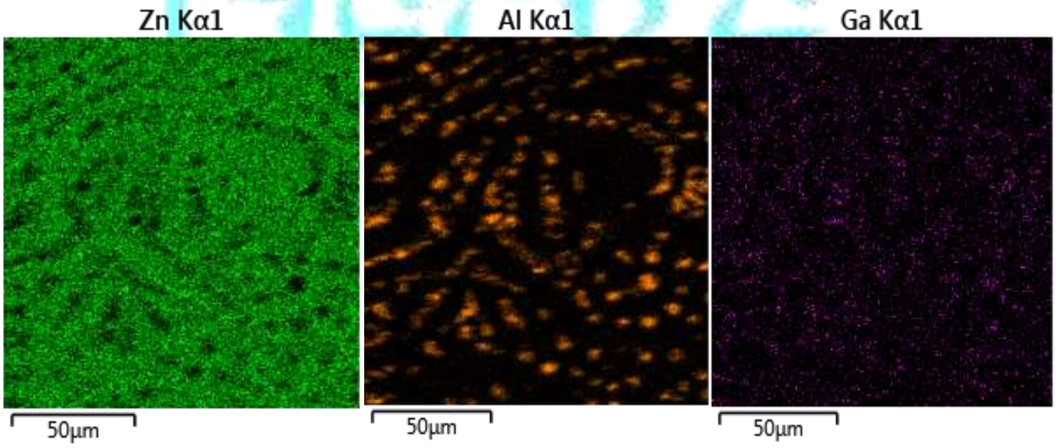

Fig.7 BSE map of elements in Zn5Al1Ga solder

For identification of phases in solder volume, the XRD analysis was performed. The diffraction record of Zn5Al1Ga solder is shown in Fig.8. The record has identified (Zn), which in solder volume occurs in the solid solution (βZn). The analysis has identified the presence of solid solution (Al) just in Zn5Al1Ga solder. The diffraction record from this zone has also identified the intermetallic phase - $Al_{0.95}Ga_{0.05}$. This means that Ga does not occur in Zn5Al1Ga solder absolutely but just in the form of intermetallic phase - $Al_{0.95}Ga_{0.05}$, eventually is dissolved in Zn or Al.

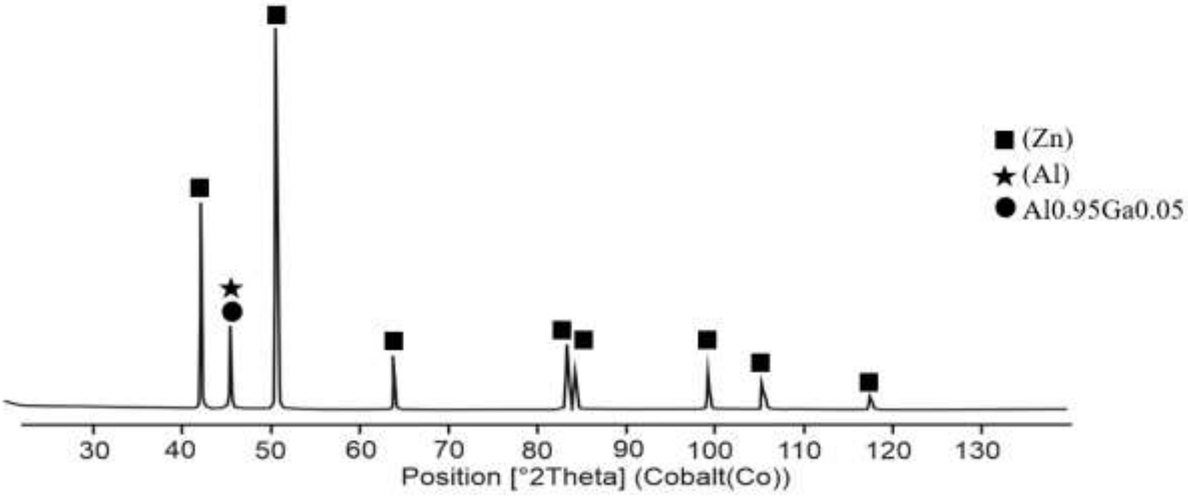

Fig.8 XRD record of Zn5Al1Ga solder

III.4 XRD analysis of fractured surface

The record from this diffraction analysis is documented in Fig. 9. The XRD analysis of Zn-Al-Ga solder has proved the presence of SiC phase and an intermetallic phase of $Al_{3,21}Si_{0.47}$.

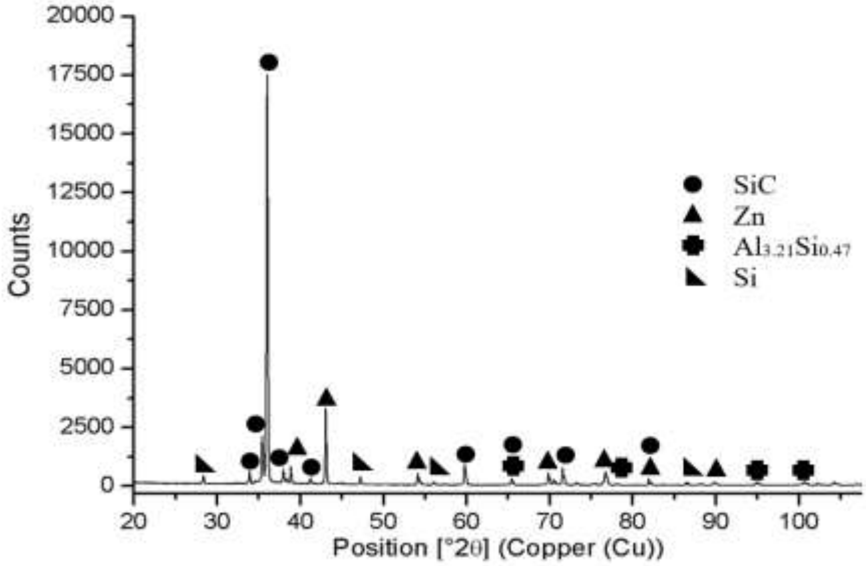

Fig.9 XRD analysis of the fractured surface of SiC/Zn5Al1Ga joint

III.5 The results of shear strength

Fig.10, where the results from shear strength test of soldered joints are documented clearly show significant effect of addition of the third element with low melting point (1 wt. % Ga).

Gallium increases the shear strength by almost 22 %. It is caused right by the Ga presence in solder volume, which significantly affects the resultant joint strength. The solder proper with Ga addition has lower strength than the eutectic Zn-Al solder, but in case of soldering it plays a significant role in reaction with the surface of SiC ceramics. This reaction results in an increase of joint strength.

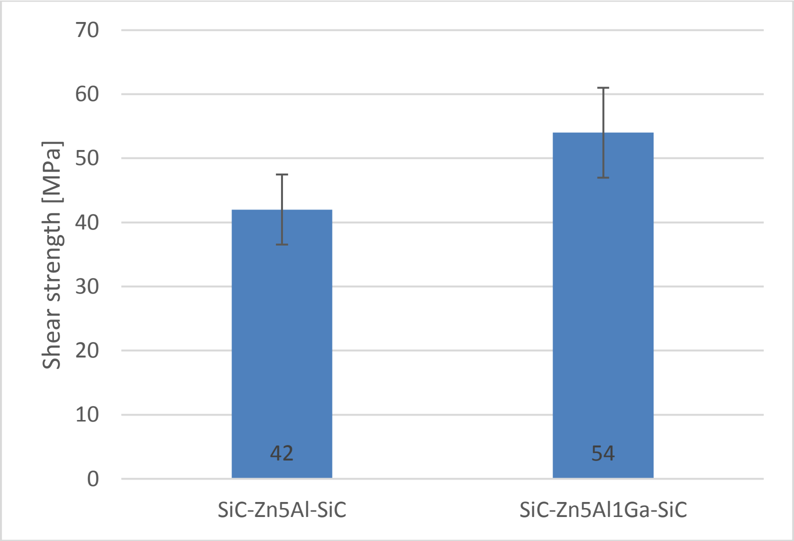

Fig.10 Shear strength of soldered joints

IV. Conclusion

This research was oriented to the study of soldering SiC ceramics by use of Zn5Al1Ga solder. The joints were fabricated by use of technology of ultrasonic soldering. The achieved results suggest the following:

- The DSC analysis of Zn5Al1Ga solder has revealed two endometric reactions at its heating. The first has occurred at the temperature of 243.3 °C and the second at 370.9 °C. Complete melting of solder occurred at the temperature of 395.5 °C, based on which the resultant soldering temperature was selected at 420 °C,
- Tensile strength of Zn5Al1Ga solder has attained the value of 83 MPa. For comparison, the Zn5Al solder has attained the value of 183 MPa,
- The solder microstructure was studied by EDX analysis, which shows the structure of Zn5Al1Ga solder. This solder consists of a solid solution (Zn) and eutectics (Zn) + (Al). Ga in solder is uniformly distributed in entire solder volume.
- For identification of phases in the solder, the XRD analysis was performed and it has identified the intermetallic phase of $Al_{0.95}Ga_{0.05}$,
- The XRD analysis performed in the boundary of SiC/Zn5Al1Ga joint has identified the intermetallic phase of $Al_{3,21}Si_{0.47}$,
- The results of shear strength measurement suggest that the SiC/Zn5Al1Ga/SiC joints have attained the value of 54 MPa.

I. Acknowledgements

This work was supported by the Agency for support of research and development, based on the contract No. APVV-17-0025 and VEGA project No. 1/0303/20: Research of joining the metallic and non-metallic materials in production of semiconductor parts.

REFERENCES

[1] WANG, H., HU, X., JIANG, X., LI, Y.: Interfacial reaction and shear strength of ultrasonically-assisted Sn-Ag-Cu solder joint using composite flux. In Journal of Manufacturing Processes, Volume 62, 2021, pp 291-301

[2] XU, Y., MA, X., XIU, Z., YAN, J.: Bonding and strengthening mechanism on ultrasonic-assisted soldering of sapphire using Sn-3.5Ag-4Al solder. In Journal of Materials Processing Technology, Volume 288, 2021

[3] YI, X., ZHANG, R., HU, X.: Study on the microstructure and mechanical property of Cu-foam modified Sn3.0Ag0.5Cu solder joints by ultrasonic-assisted soldering. In Journal of Manufacturing Processes, Volume 64, 2021, pp 508-517

[4] WU, B., LENG, X., XIU, L., YAN, J.: Microstructural evolution of SiC joints soldered using Zn-Al filler metals with the assistance of ultrasound. In Ultrasonics Sonochemistry, Volume 44, 2018, pp 280-287

[5] ZHANG, X., LI, D., WANG, L., WAN, CH., SONG, L., XIAO, Y.: Ultrasonic brazing of Zr-based bulk metallic glass and 1060 Al alloy using Zn-3Al filler metal. In Journal of Advanced Joining Processes, Volume 5, 2022

[6] HAQUE, A., WON, Y. S., HASEEB, A.S.M.A., MASJUKI, H.H.: Investigations on Zn-AI-Ge Alloys as High Temperature Die Attach Material. In Electronic System-Integration Technology Conference (ESTC) 3rd, 2010

[7] NAGAOKA, T., MORISADA, Y., FUKUSUMI M., TAKEMOTO, T.: Selection of soldering temperature for ultrasonic-assisted soldering of 5056 aluminum alloy using Zn–Al system solders. In Journal of Materials Processing Technology, vol. 211, 2011, pp. 1534-1539. DOI: 10.1016/j.jmatprotec.2011.04.004

[8] CHENG, F., GAO, F., WANG, Y., WU, Y., MA, Z., YANG, J.: Sn addition on the tensile properties of high temperature Zn–4Al–3Mg solder alloys. In Microelectronics Reliability, vol. 52, 2012, pp. 579-584. DOI: 10.1016/j.microrel.2011.10.003

[9] HAQUE, A., LIM, B.H., HASEEB, A.S.M.A., MASJUKI, H.H.: Die attach properties of Zn-Al-Mg-Ga based high-temperature lead-free solder on Cu lead-frame. In Journal of Material Science: Materials in Electronics, Vol. 23, 2012, pp. 115-123. DOI: 10.1007/s10854-011-0511-x

[10] TAKAKU, Y., FELICIA, L., OHNUMA, I.: Interfacial Reaction Between Cu Substrates and Zn-Al Base High-Temperature Pb-Free Solders. In Journal of Electronic Materials, Vol. 37, Is. 3, 2008, pp. 314-323. DOI: 10.1007/s11664-007-0344-9

[11] ALIBABAIE S., MAHMUDI, R.: Microstructure and Creep Characteristics of Zn-3Cu-xAl Ultra High-Temperature Lead-Free Solders. In Materials and Design, Vol. 39, 2012, pp. 397-403. DOI: 10.1016/j.matdes.2012.03.005

[12] ZENG, G., MCDONALD, S., NOGITA, K.: Development of High-Temeprature Solders: Review. In Microelectronics Reliability, Vol. 52, 2012, pp. 1306-1322. DOI: 10.1016/j.microrel.2012.02.018

[13] SHIMIZU T., ISHIKAWA, H., OHNUMA, I., ISHIDA, K.: Zn-Al-Mg-Ga Alloys as Pb-Free Solder for Die-Attaching Use. In Journal of Electronic Materials, Vol. 28, No. 11, 1999

[14] KOLENAK, R., KOSTOLNY, I., CICKA, R.: *Research of Fluxless Soldering of High-Purity Aluminium with Solders Type Zn-Al.* In Advanced Materials Research, Vol. 905, 2014, pp. 132-136: DOI: 10.4028/www.scientific.net/AMR.905.132