International Journal of Scientific and Technical Research in Engineering (IJSTRE)
www.ijstre.com Volume 6 Issue 6 ǁ Nov-Dec 2021.
ISSN: 2581-9941

Research on soldering of ceramic materials by hybrid laser/ultrasound technology

Igor Kostolny[1], Tomas Melus[1*], Roman Kolenak[1], Paulina Babincova[1] and Michal Simek[2]

[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)*
[2]*(The First Welding Company Inc., Kopcianska No. 14, 851 01 Bratislava 5, Slovak Republic)*
tomas.melus@stuba.sk

Abstract: *This work deals with the study of soldering ceramic materials by application of a hybrid technology – laser/ultrasound. The wettability of Sn5Sb3Ti solder on the ceramic substrates as Al_2O_3 and SiC and microstructure of fabricated joints was assessed. The wettability of this solder on Al_2O_3 ceramics at temperature of 710 °C attained the values from 35 to 40°. On the SiC substrate and at the same temperature, the wetting angles from 22 to 25° were attained. The microstructure in the boundary of soldered joints was analysed by use of SEM/EDX analysis. The planar analysis of elements in joints between the ceramic substrates and solder has revealed the presence of higher concentration of Ti. Wetting of ceramic materials was ensured by bonding of titanium with the surface of substrates via formation of a very thin reaction layer.*

Keywords - *soldering, Al_2O_3, SiC, ceramic, ultrasonic soldering, laser beam, microstructure.*

I. INTRODUCTION

The Al_2O_3 or SiC ceramics are used in many industrial branches, as for example the space, automotive and aviation industries. These are applied as components mainly in electronics or power engineering owing to their remarkable mechanical properties and low coefficient of thermal expansivity.

The ceramic materials in electronic power equipment are usually joined by use of solders. However, the commercial solders do not wet these materials, therefore the so-called active soldering alloys, alloyed with active elements are used [1].

The active elements play at present a significant role in soldering alloys. Here belong mostly the elements as Ti, V and/or lanthanides. These elements are added to solder in order to ensure the wetting of ceramics during soldering process [1, 2, 3, 4].

The solders containing active elements react with the surface of ceramic material during soldering process, what leads to formation of a reaction layer on the solder and substrate boundary.

Application of a combined technology – laser/ultrasound has proved to be a very suitable technology for soldering ceramics. The ultrasonic vibrations applied on a molten solder cause a rapid formation of cavitation microbubbles, what results in formation of local zones with extremely high temperature and pressure. These subsequently remove the thin oxide layer and improve thus the solder wettability. This results in grain refining and thus also the physical-chemical interactions between the substrate and solder are significantly affected [8, 14].

The most recent studies have shown that ultrasonic soldering supports the formation of reaction layer. Thus, the wetting of a ceramic material is ensured [5, 6, 7, 8].

Ultrasonic soldering is suitable process for joining hard-to solder materials. Ultrasonic soldering is mostly employed in many fields of fluxless soldering due to its capability to fabricate the joint by a direct method without the need for previous surface treatment [8, 12, 13].

The authors [11] have dealt with the wettability study of Al_2O_3 ceramics by use of an active solder type Sn3,5Ag4Ti. The experiment was performed in vacuum at a high activation temperature of 800 to 900 °C. Titanium activation in solder necessitates the achieving of high soldering temperature.

Another procedure how to achieve titanium activation in solder consists in activation by ultrasonic power, what subsequently leads to local temperature increase. The solder type Sn3,5Ag4Ti contains just a small amount of active titanium element, but even so it is capable to wet the surface of Al_2O_3 ceramics.

The authors in work [15] have studied the wettability of SiC ceramics by an active solder type Sn-Ti. The experiment was realised in an oven at high temperature from 750 to 1000 °C. The highest wettability value – 20° was achieved with this solder at the temperature of 1000 °C.

Laser soldering is at present applied for electronic equipment on printed circuit boards, owing to its unique properties as the local and contactless heating, rapid temperature increase and drop and easy automation, when compared to the course of soldering with remelting [16, 17].

This study deals with soldering Al_2O_3 and SiC ceramics by use of Sn-Sb-Ti – based solder. The work is oriented to quality assessment of soldered joints by application of a selected solder and the hybrid soldering technology – laser/ultrasound. The analysis is oriented to qualitative assessment of solder wettability on the ceramic substrates and the boundaries of soldered joints.

II. EXPERIMENTAL

Soldering is defined as a way of metallurgical joining of metallic parts by a molten solder, whereby the joined surfaces are not molten down, but just wetted by the solder used.

For mutual joining of solder and ceramic material it is necessary that the solder would wet the surface of parent substrate: wettability is characterised as a capability of molten solder to adhere to the clean surface of joined substrate at a certain temperature.

The quantitative criterion is exerted by the contact angle, which is included by the tangent of solder surface with the surface of a selected substrate in the point of their contact.

The measurement of contact angle by the goniometric method was realised on the specimens of ceramic materials as Al_2O_3 and SiC, on which the solder in form of a cube with the edge dimension of 4 mm was laid. Fig. 1 shows the layout of substrate and solder. The substrates were in the form of discs in diameter of 15 mm and 3 mm in thickness. The chemical composition of used solder is given in Table 1. In the soldering process realised by laser/ultrasound technology, the shielding gas argon was applied.

Table 1 Chemical composition of solder

Solder designation	**Chemical composition of solder [wt %]**
Sn5Sb3Ti	92% Sn, 5% Sb, 3% Ti

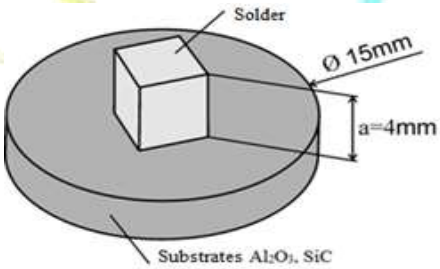


Fig. 1 Schematic representation of substrate and solder

In order to achieve the solder activation the equipment type Hanuz UT2-ultrasonic transducer was applied using a piezoelectric system with a titanium sonotrode in diameter of Ø3 mm. Table 2 gives the ultrasound parameters of this equipment. Heating of studied solder was realised by use of a solid-state laser type YLR 4500. Maximum power of this laser is 4500W, with the wavelength of 1.06 µm. The experiment was performed with a laser head type Precitec YW50, with the diameter of optical fibre of 0.3 mm.

Table 2 Ultrasound parameters

Ultrasound power	[W]	400
Work frequency	[kHz]	100
Amplitude	[µm]	2
Soldering temperature	[°C]	600 – 720
Time of ultrasound acting	[s]	5

The parameters of laser soldering for the actual soldered joints are given in Table 3. The set value of focal distance of laser head was z (-42,5). Heating was in experiment realised through a graphite jig, into which the substrate and solder were inserted. In order to achieve the desired temperature, laser beam made the cruciform movements around the periphery of a graphite jig. During soldering and subsequent cooling down, argon protection against the oxidation of molten solder in the air was applied.

Table 3 Parameters laser equipment

Specimens	Shielding gas	Power (W)	Heating temperature (°C)	Numer of circulation
Al_2O_3/Sn5Sb3Ti	Argon (F10)	750W	710 °C	30
SiC/Sn5Sb3Ti	Argon (F10)	750W	710 °C	30

During heating the laser equipment rotated in close vicinity of substrate, without a direct contact with the substrate. This way, the graphite jig was heated and subsequently transferred the heat to the substrate and solder. After melting down of soldering alloy an ultrasonic sonotrode was immersed to the melt by an automatic movement for the time of 5 second. Fig.2 shows the schematic representation of soldering process by the laser/ultrasound technology.

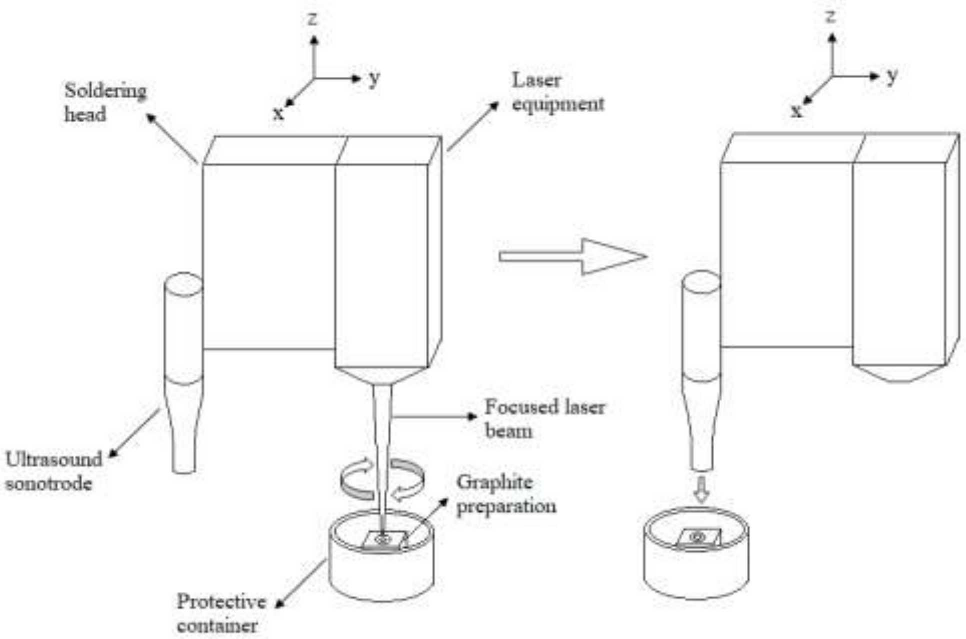


Fig. 2 Schematic representation of soldering by laser/ultrasound process

The resultant analysis of boundary between the solder and substrate was performed by the electron scanning microscopy (SEM) on equipment type TESCAN VEGA 3. The qualitative and quantitative chemical analysis was performed on equipment type JOEL 7600 F, provided with an X-ray microanalyser type Microspec WDX-3PC.

III. Results

III.1 Measurement of solder wettability on the ceramic substrates

It is known at present that the ceramic materials are not wettable by the commercial solders. Any analysed solder must therefore contain an element, which is then chemically active to an actual type of ceramics. The analysed solder type Sn5Sb3Ti contains titanium and antimony. Titanium exerts a high affinity to oxygen, carbon or silicon and therefore it can be at suitable conditions bound to some component of ceramics. The measured wettability results, attained by the goniometric method applied for determination of contact wetting angles, are documented in Fig. 3a) and compared on a graph in Fig. 3b).

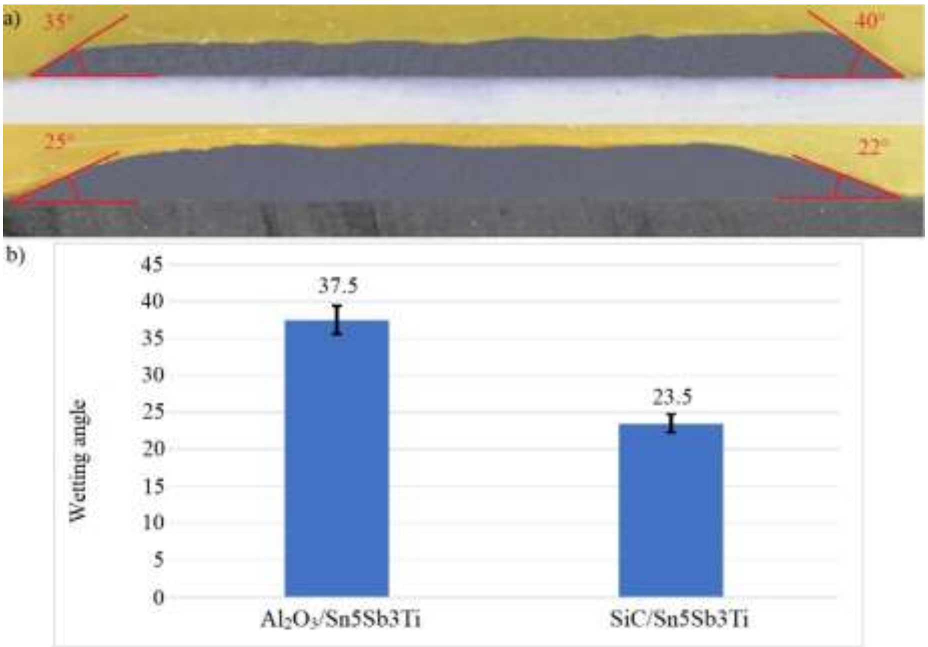


Fig. 3 Measurement of wetting angle of Sn5Sb3Ti solder on ceramic substrates
a) results of goniometric method of measurement; b) comparison of wetting angles

The Sn5Sb3Ti solder exerted very low values in wettability measurements and therefore very good results of wettability were achieved. The best measured value of contact angle was achieved on SiC substrate with the average value of 23.5 °. The wettability on Al_2O_3 substrate has attained the average wetting angle of 38.5 °.

The results from study [11] provided remarkable findings, since in comparison with the wettability results of solder type Sn-Ag-Ti on Al_2O_3 substrates, the resultant wettability was lower by approximately 37.5°. In the case of Al_2O_3 solder and used solder type Sn-Ti mentioned in work [16], the overall wettability at the temperature of 900°C in vacuum was by approximately 10° higher. Ultrasound assistance thus provides significantly lower values. Such low wettability values are not currently observed with the ceramic substrates.

The chemical reactions taking place between the molten solder and ceramics are necessary precondition for achieving wetting.

III.2 Analysis of boundaries in soldered joints

Fig.4 shows the results of planar EDX analysis of boundary in Al_2O_3/Sn5Sb3Ti joint. The continuous Ti layer (yellow colour) has decomposed in the joint boundary during soldering. The titanium bond in the boundary is formed owing to high affinity of titanium to oxygen, when the titanium oxides are bound to the surface of Al_2O_3 ceramics. The substrate wetting and bond formation are significantly affected by Ti, which forms the reaction products, owing to its reactions in the boundary.

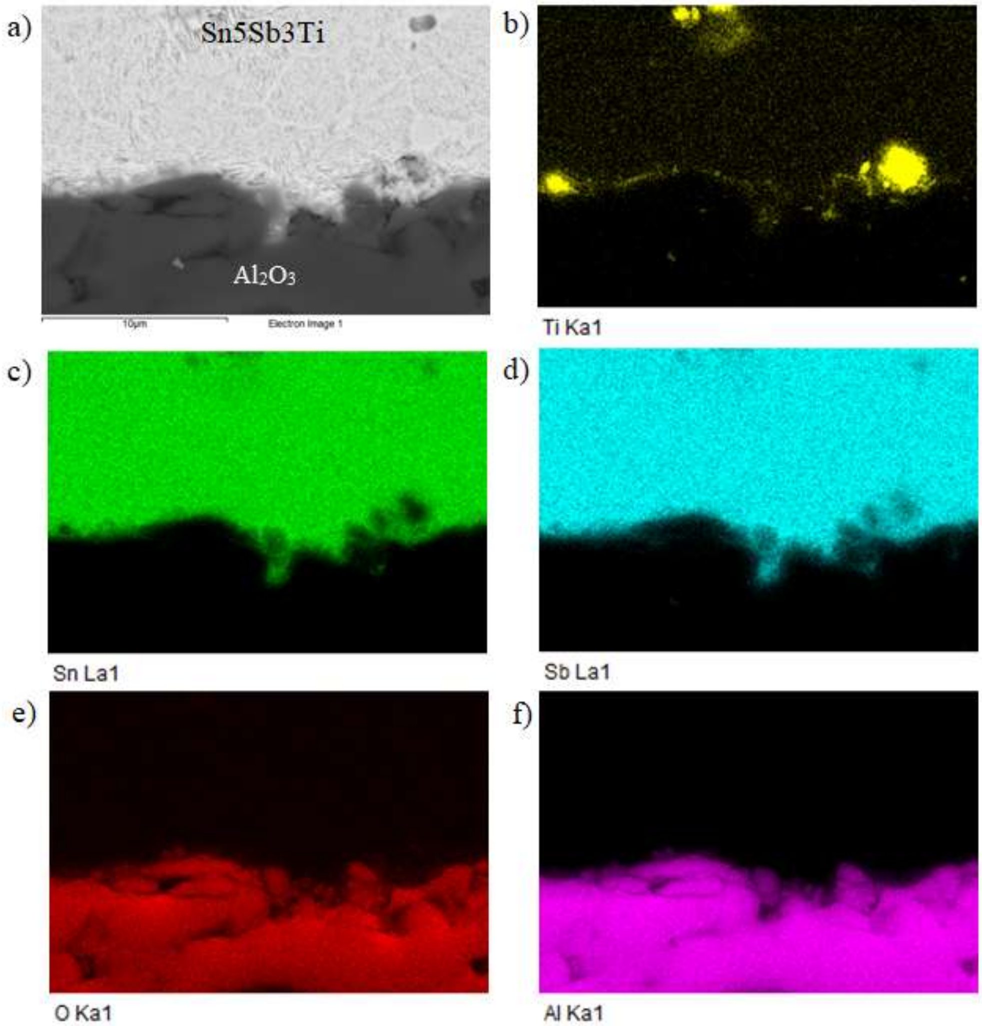


Fig. 4 Planar EDX analysis of the boundary in Al_2O_3/Sn5Sb3Ti joint
a) joint boundary; b) Ti content

The point analysis allowed to determine the proportion of elements in the close boundary between the used solder and substrate. The distribution of elements in percentage grouping in the joint boundary may be seen Fig. 5. Measurement in spectra 1 and 2 identified the chemical composition consisting mainly of Al, Ti, Sn and Sb. The point analysis has captured the Ti presence, mainly in the zone of analysed boundary and in the particles contained in solder volume. Titanium in the joint boundary was bound with the oxygen atoms from ceramics and has formed a very thin reaction layer in the thickness of 1 to 3 µm.

Spectrum	Al [wt. %]	Ti [wt. %]	Sn [wt. %]	Sb [wt. %]
Spectrum 1	7.81	1.62	86.81	3.76
Spectrum 2	3.68	0.12	92.04	4.17
Spectrum 3	0.14	35.13	25.04	39.69
Spectrum 4	0.25	16.70	51.65	31.40
Spectrum 5	0.26	0.00	95.37	4.37
Spectrum 6	1.72	0.25	94.68	3.35

Fig. 5 Point EDX analysis of boundary in Al_2O_3/Sn5Sb3Ti joint

Analysis of planar distribution in the boundary of SiC ceramics and Sn5Sb3Ti solder is shown in Fig. 6. Titanium presence is marked by yellow colour. The continuous titanium layer in boundary is less observable by this analysis, but its distribution to boundary is evident. The EDX analysis has identified also a big Ti phase close in boundary vicinity, reaching to the solder volume.

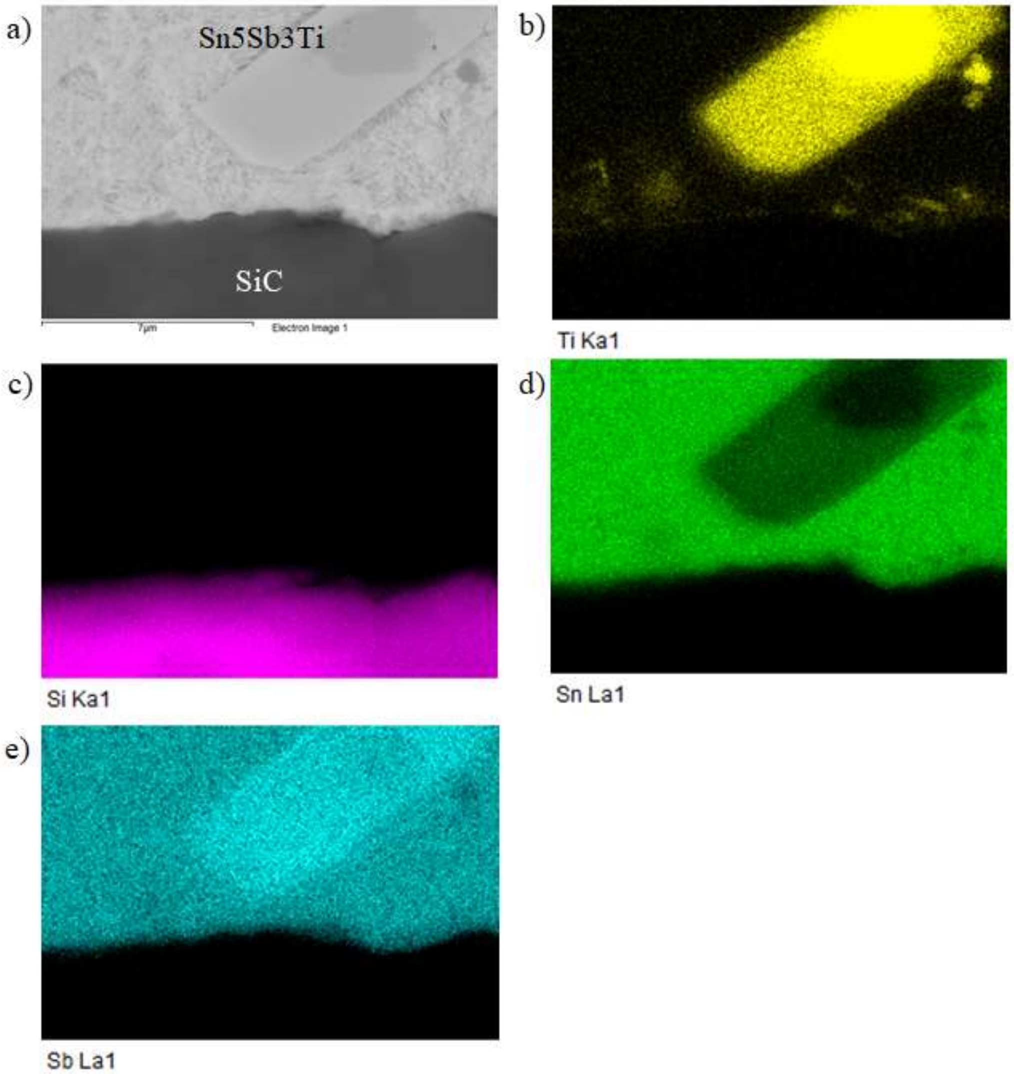
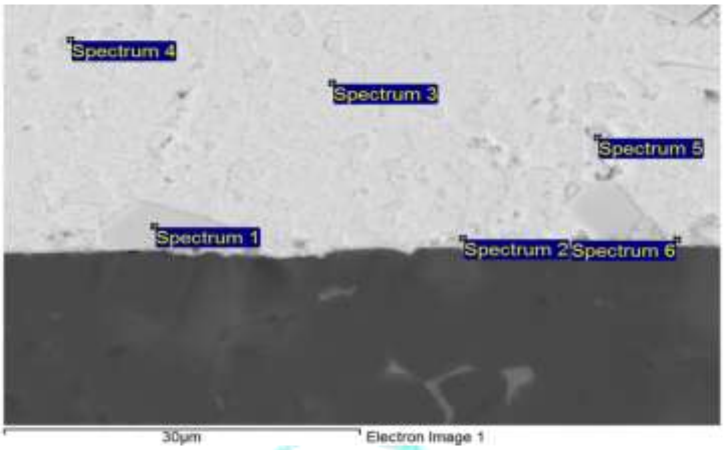


Fig. 6 Planar EDX analysis of the boundary in SiC/Sn5Sb3Ti joint
a) joint boundary; b) Ti content

The point EDX analysis determined the proportion of elements on the close boundary between the used solder and substrate. The distribution of elements in percentage grouping in the joint boundary may be seen in Fig. 7. The analysed boundary marked by spectra 1, 2 and 6 has identified the significant effect of Ti on bond formation.



Spectrum	Si [wt. %]	Ti [wt. %]	Sn [wt. %]	Sb [wt. %]
Spectrum 1	0.52	17.01	48.79	33.68
Spectrum 2	3.71	1.46	88.96	5.87
Spectrum 3	-	-	94.48	5.52
Spectrum 4	-	-	94.97	5.03
Spectrum 5	0.44	-	93.91	5.65
Spectrum 6	10.09	4.54	77.65	7.72

Fig. 7 Point EDX alalyis of boundary in SiC/Sn5Sb3Ti joint

IV. CONCLUSION

The work deals with the study of soldering ceramic materials as Al2O3 and SiC by use of Sn5Sb3Ti solder. Soldering was realised by the hybrid technology of laser/ultrasound. The achieved results may be characterised as follows:

- The wettability of solder type Sn5Sb3Ti studied on Al_2O_3 and SiC substrates at the temperature of 710 °C (provided by laser beam in combination with ultrasound assistance) has attained the values from 22 to 40 °.
- In assessment of planar analysis of elements in the joints between the ceramic materials and Sn5Sb3Ti solder, the presence of higher concentration of Ti was observed, which formed a reaction layer and ensured thus the wetting of both ceramics.
- The point EDX analysis performed on the close boundary of Al_2O_3/Sn5Sb3Ti joint has identified mainly the Ti, Al, Sb and O elements. It is supposed that the high affinity of titanium to oxygen in the process of hybrid soldering has ensured the reaction of titanium with the surface of ceramics and subsequently formed a thin TiO layer in the thickness ranging from 1 to 3 µm.
- The reaction of Ti in solder with the surface of SiC substrate has ensured the wetting of this ceramics. The EDX analysis has clearly identified Ti presence on the joint boundary. It is supposed that bond formation is affected by the reaction of Ti with Si and C.

The next research of these joints will be oriented to identification of the strength characteristics and the analysis of fractured surfaces of soldered joints in dependence on the applied soldering parameters.

V. Acknowledgements

This work was supported by the Slovak Research and Development Agency under the contract no. APVV-17-0025. The paper was prepared also with the support of the VEGA 1/0303/20 project: Research of joining the metallic and ceramic materials in production of power semiconductor.

REFERENCES

[1] JUNG, Do-Hyun, Ashutosh SHARMA a Jae-Pil JUNG. Influence of dual ceramic nanomaterials on the solderability and interfacial reactions between lead-free Sn-Ag-Cu and a Cu conductor: Journal of Alloys and Compounds. *Journal of Alloys and Compounds* [online]. [cit. 2021-9-29]. Dostupné z: doi:https://doi.org/10.1016/j.jallcom.2018.02.017.

[2] Lovro Gorjan, Gurdial Blugan, Manfred Boretius, et al. Fracture behavior of soldered Al2O3 ceramic to A356 aluminum alloy and resistance of the joint to low temperature exposure. *Materials & Design* [online]. 2015, , 889-896 [cit. 2021-10-4]. ISSN ISSN 0264-1275. Dostupné z: https://www.sciencedirect.com/science/article/pii/S0264127515304779

[3] CHANG, S.Y. HUNG, Y.T. CHUANG, T.H. *Joining alumina to inconel 600 and UMCo-50 superalloys using an Sn10Ag4Ti active filler metal.* J Mater Eng Perform, 12 (2003), pp. 123-127

[4] S. Kang, J.H. Selverian.. *Interactions between Ti and alumina-based ceramics.* J Mater Sci, 27 (1992), pp. 4536-4544

[5] Q. Gu, Y.X. Wang, Y.D. Cui, X. Chen, K. Tao. *Interfacial reaction of Ti and sapphire.* Wuli Xuebao/Acta Phys Sinica, 45 (5) (1996)

[6] J.J. Stephens, F.M. Hosking, T.J. Headley, P.F. Hlava, F.G. Yost. *Reaction layers and mechanisms for a Ti-activated braze on sapphire.* Metall Mater Trans A, Phys Metall Mater Sci, 334 (12) (2003), pp. 2963-2972

[7] A. Xian. *Joining of sialon ceramics by Sn-5 at% Ti based ternary active solders.* J Mater Sci, 32 (1997), pp. 6387-6393

[8] RASHIDI, Roxana a Homam NAFFAKH-MOOSAVY. *Metallurgical, Physical, Mechanical and Oxidation Behavior of Lead-Free Chromium Dissolved Sn-Cu-Bi Solders. Journal of Materials Research and Technology [online]. 2021, 31.6.2021 [cit. 2021-6-2].* ISSN 2238-7854. Available from: https://www.sciencedirect.com/science/article/pii/S2238785421004956

[9] LANIN, V.L. Ultrasonic soldering in electronics. Ultrason Sonochem, 8 (2001), pp. 379-385, 10.1016/S1350-4177(01)00065-7

[10] TAN, A.T. TAN, A.W. YUSOF F. Effect of ultrasonic vibration time on the Cu/SnAgCu/Cu joint soldered by low-power-high-frequency ultrasonic-assisted reflow soldering. Ultrason - Sonochemistry, 34 (2017), pp. 616-625, 10.1016/j.ultsonch.2016.06.039

[11] DREVO, EP. NIMMO, KL. *In search of new unleaded electronic solder.* Elektron. Mater. 23 (8) (1994), str. 709 – 713

[12] KOLEŇÁK, R. ŠEBO, P. PROVAZNÍK, M. a KOLEŇÁKOVÁ, M. *Shear strength and wettability of active Sn3.5Ag4Ti(Ce,Ga) solder on Al_2O_3 ceramics*. 7, August 2011, 32, Pages 3997-4003. Dostupné z: doi:https://doi.org/10.1016/j.matdes.2011.03.022

[13] KM Hafez, M. Naka. Mechanism of ultrasonic irradiation on joining of alumina/copper. Trans. JWRI, 31 (2002), s. 177 – 180

[14] WY Yu, SH Liu, XY Liu, MP Liu, WG Shi. Interface reaction in ultrasonic vibration-assisted brazing of aluminum to graphite using Sn–Ag–Ti solder foil. J. Mater. Proces. Technol, 221 (2015), s. 285 - 290

[15] Koleňák, R.; Kostolný, I.; Drápala, J.; Sahul, M.; Urminský, J. Characterizing the Soldering Alloy Type In–Ag–Ti and the Study of Direct Soldering of SiC Ceramics and Copper. Metals 2018, 8, 274.

[16] S. Reinl,. *Diode Lasers used in Plastic Welding and Selective Laser Soldering – Applications and Products*, Physics Procedia,Volume 41,2013,Pages 234-240,ISSN 1875-3892, https://doi.org/10.1016/j.phpro.2013.03.074.(https://www.sciencedirect.com/science/article/pii/S1875389213000886)

[17] Koleňák, R., Chachula, M., Šebo, P. and Koleňáková, M. (2011), "Wettability and shear strength of active Sn2Ti solder on Al2O3 ceramics", Soldering & Surface Mount Technology, Vol. 23 No. 4, pp. 224-228.

[18] A. Kar, S. Mandal, R. N. Ghosh, T. M. Ghosh, and A. K. Ray, " Role of Ti diffusion on the Formation of Phases in the Al2O3–Al2O3 Brazed Interface," J. Mater. Sci., 42 5556– 5561 (2007).

[19] LEE, Hwa-Teng, Heng-Sheng LIN, Cheng-Shyan LEE a Po-Wei CHEN. Reliability of Sn–Ag–Sb lead-free solder joints. *Materials Science and Engineering* [online]. 2005, 2005, Pages 36-44 [cit. 2021-9-29]. ISSN ISSN 0921-5093. Dostupné z: doi:https://doi.org/10.1016/j.msea.2005.07.049