Structure and Properties of Dielectric Coatings Based on Fusible Glass-Ceramic Materials

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Abstract* – *Structure and thermal properties of the dielectric coatings on the basis of glass-ceramic system PbO-ZnO-B_2O_3-SiO_2-Al_2O_3-BaO (sealant) on a stainless steel substrate were studied. The advantages and disadvantages of these surface layers in comparison with dielectric surface layers applied according to the method of magnetron sputtering are shown.

Key words – flat heating element, surface engineering, dielectric coatings, glass crystalline material, thermal conductivity, thermal diffusivity, heat treatment.

I. Introduction

Application of electric current as a source of thermal energy in the conditions of increasing scarcity of natural energy is becoming more important. The development of environmentally friendly electric heaters with low inertia for space heating, trains and municipal electric (cabins and carriages for trains, trams, trolley buses) is one of energy conservation problems. At present, the production technology of flat heaters is expensive and with low productivity. This technology based on multiple screen printing method followed by complicated heat treatment. Moreover, for the production of dielectric and resistive pastes were used precious metals. The principle scheme of a flat heating element structure is shown in Fig. 1.

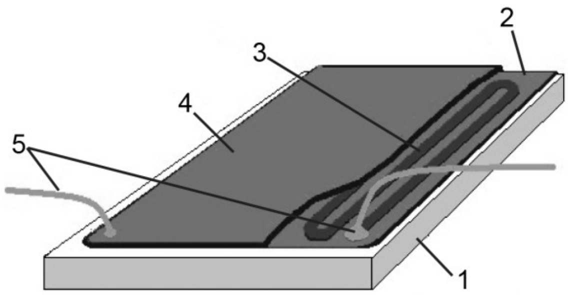


Fig. 1. Principle scheme of a flat heating element [1]:
1 – substrate; 2 – dielectric layer; 3 – resistive element;
4 – protective shell; 5 – contact electrodes

This work focuses on the development of alternative production technologies of dielectric layer (Fig.1) for flat heating elements by methods of surface engineering with using new dielectric pastes based on glass ceramics system PbO-ZnO-B_2O_3-SiO_2-Al_2O_3-BaO (sealant) with minimal proportion of precious metals or without it, on substrate with stainless steel. The thermal conductivity and thermal diffusivity of the three glass-ceramic surfaces which manufactured from SC 90–1 and SC 100–1 powders, were investigated. The obtained results were compared with the properties of the substrate. SC 90–1 and SC 100–1 powders, were investigated. The obtained results was compared with the properties of the substrate.

II. Experimental part

To obtain radically new dielectric layers on flat heating elements, there were chosen three brands of sealants powder: SC 90–1, SC 88 and SC 100–1 (Table. 1). Before applying to the surface of the substrate, the powder was added to 30-40 % of Butyl acetate and 2-6 % of nitrocellulose varnish. After careful stirred during 3–4 hours, the resulting slurry was applied to the surface of the stainless steel plate 50*50 mm.

TABLE 1

THE CHEMICAL COMPOSITION OF INITIAL POWDERS

Oxide	Chemical composition, wt. % (± 0.2 %)		
	SC 100–1	SC 90–1	SC 88
PbO	75.5	75.3	75.1
ZnO	12	11.6	11.2
B_2O_3	8.4	8.5	9.4
SiO_2	2.1	2.1	1.9
Al_2O_3	2	0.8	–
BaO	–	1.7	1.88

Obtained surface of the samples were dried in a furnace chamber at 70 ºC. The final step of the creation of dielectric coating was complex heat treatment during which crystallization occurs. The scheme of heat treatment mode for the crystallization of dielectric layer shown in Fig. 2.

As a result of painstaking research, we found the optimum heat treatment mode of three sealents which consists of two stages:

– the first stage of heat treatment – annealing at t_1 = 380 °C during 45 minutes to forme of the crystallization centers. Heating from the room temperature to 400 °C carried out at the speed of 4 °C/min. Temperature level was maintained by thermostat within ± 5 °C;

– the second stage of heat treatment – was annealing at t_2 = 440 ° C, during which the maximum speed of crystal growth implemented. The duration of the second stage annealing corresponds to the required time for full volume crystallization of the glass layer. Increasing the temperature from 380 °C to 440 °C is carried out at the speed of 4 °C/min. At 440 ± 5 °C system "substrate - coating" lasted by 60 min. Then the obtained dielectric coating was spontaneous cooled to 90 °C.

The oxides and nitrides of Mg, Al, Ti layers were deposited by the modes (Tab. 2), differing by the process duration (τ), the substrate bias potential (E), the pressure (P) and the current plasma arc (I). To ensure a better adhesion of the coatings a pre-heating of the substrate by using a heater installed directly into the reaction chamber of the ion-plasma system was carried out. In addition, the final cleaning of the substrate in a stream of the argon plasma by using the helicon source in the "column" regime was carried out for 30 minutes.

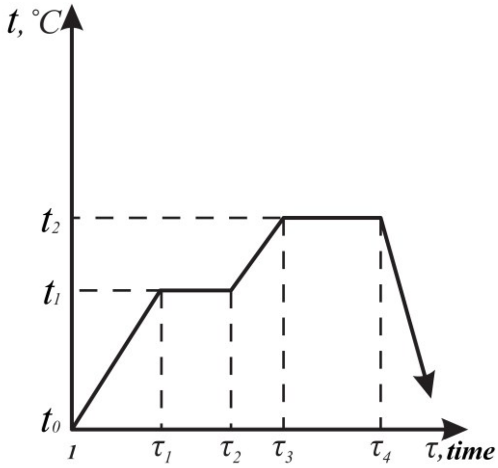


Fig. 2. Scheme of heat treatment mode
for the crystallization of dielectric layer [2]

TABLE 2
REGIMES OF ION-PLASMA SPUTTERING

Oxide	Substrate	τ, min	P, Pa	E, V	I, A
AlN	40X13	40	5,9	-60	40
MgO	AMg2	20	1,9÷2,6	-60	32

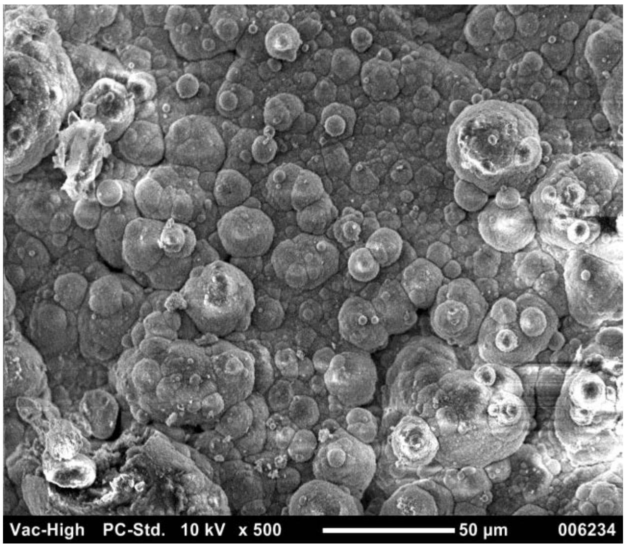
The thickness of the dielectric coating was determined by metallographic microscope Mikrotex MMT-14Cz with application software «ToupView». The study of the structure of the formed surface layers was carried out by the scanning electron microscope JSM-6490LV (JEOL, Japan), with an analytical console for the element analysis (INCA Energy – Oxford). Research of thermo-kinetic properties of obtained dielectric coatings was carried out on Laser-Flash-Apparatus device - LFA 427.

III. Results and Their Discussion

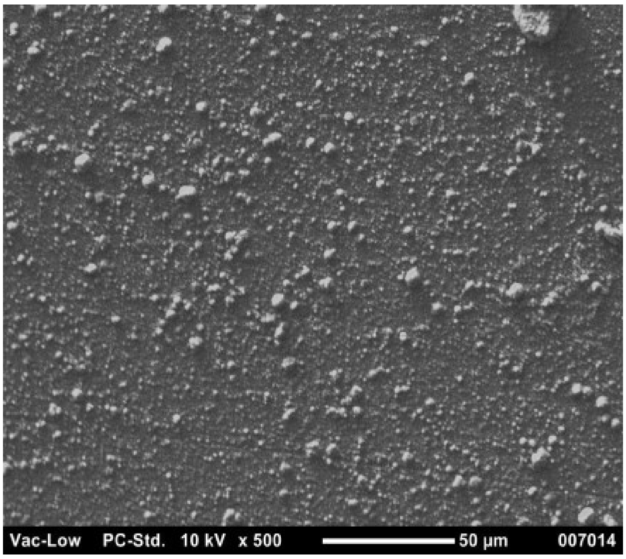
The method of making a dielectric coating of pastes and suspensions is an incredibly productive and low-cost one as compared to other methods.

At the same time, the insulating coating which we obtained with the help of magnetron sputtering of oxides and nitrides in vacuum allowed us to obtain high values of adhesion and electro-physical characteristics [3,4]. On the other hand, the application of this method does not allow us to obtain a great number of articles because the working chamber is small in size.

Investigation of the formed layers surface topology by the electronic microscopy method shows a difference in their structure. The MgO layer consists of round-shape grains with different degrees of dispersion. Grain formation occurs on the substrate surface with ions, atoms and droplet fractions of magnesium. Therefore, the size of the formed grains is widely fluctuated. It is possible that grain growing is carried out by an “island” mechanism where the formed grain is the basis for the formation of new centers of crystallization and their subsequent growth (fig. 3, a). Thus, the newly grown grains are formed in clusters of different sizes. The high density of the structure and uniformity across the surface area of the AlN layer surface was found by analyzing its microtopology. High values of the substrate bias potential and pressure during the regime of ion-plasma sputtering contributed to increasing the aluminum ions rate, the grinding the structural constituents and a qualitative forming of the layer structure (fig. 3, b).



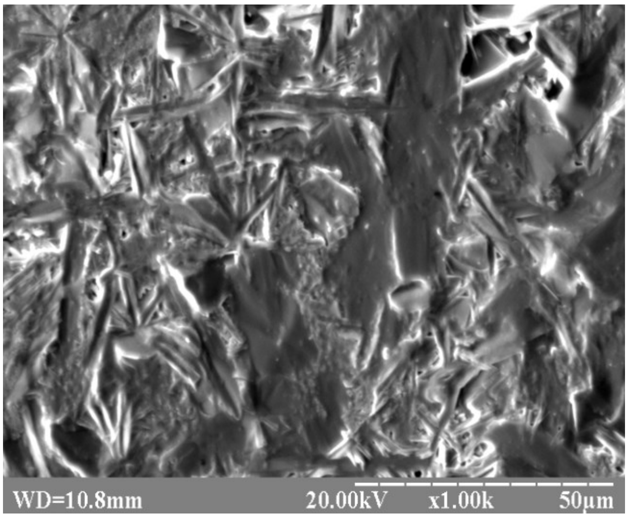
a



b

Fig. 3. SEM surface microstructure of the dielectric layers:
a – MgO, b – AlN

During the microstructure study of of the dielectric coatings on the basis of glass-ceramic system PbO-ZnO-B_2O_3 detected the dendritic structure (fig. 4, a,b,c) which indicates the successful crystallization process of formed coatings. Such coatings will have the high parameters of dielectric strength. as opposed to amorphous glass-ceramic materials.



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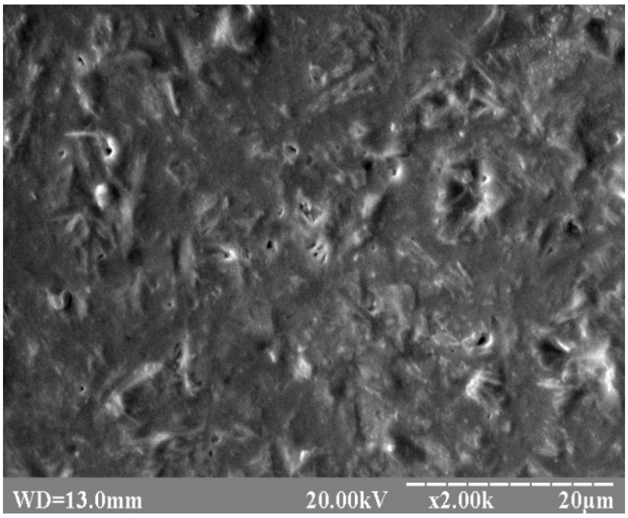

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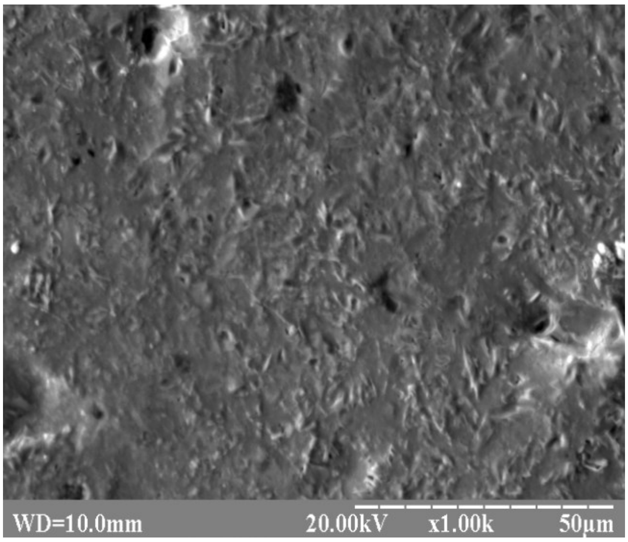
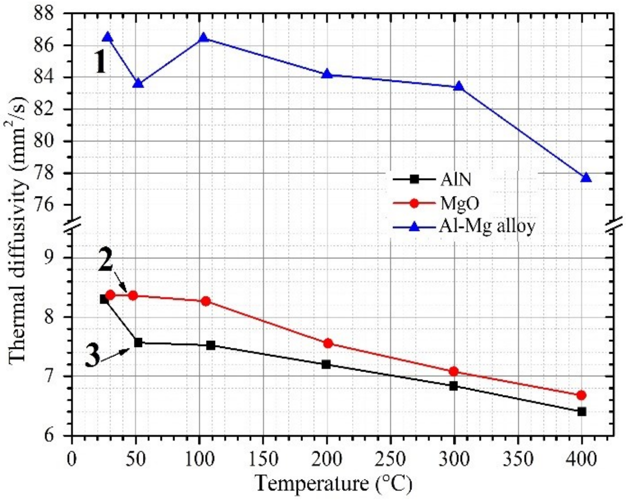


c

Fig. 4. SEM surface microstructure of the dielectric layers:
a – SC 100–1, b – SC 90–1, b – SC 88

Thermal diffusivity and heat conductivity over the 0 to 500 °C temperature range were investigated in order to compare thermo-kinetic properties of obtained dielectric layers (sealants and oxide/nitride layers) and substrate. First, these studies were implemented to oxide layers obtained by magnetron plasma sputtering. The values of heat conductivity and thermal diffusivity of AlN and MgO on the Al-Mg alloy substrate are shown in Fig. 5.



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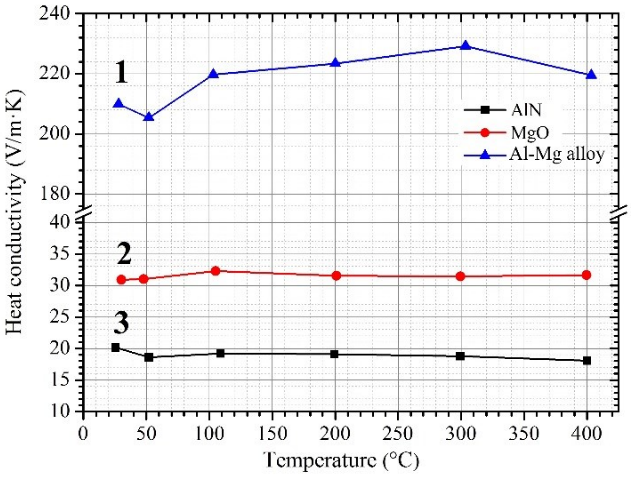


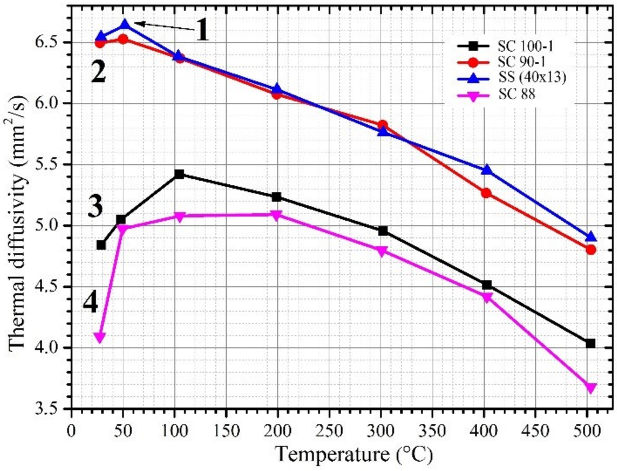
b

Fig. 5. Thermal diffusivity (a) and heat conductivity (b) of dielectric layers MgO (2) and AlN (3) on substrate of Al-Mg alloy (1)

Obtained surface layers on the basis of SC 90–1, SC 88 and SC 100-1 sealants posses better likeness of thermo-physical properties with the substrate stainless steel substrate (Fig. 6). The discontinuity of thermal expansion in the system "AlN/MgO – Al-Mg alloy substrate" may cause an emergence of structural blemishes during the "cooling-heating" cycles of surface. This leads to the deterioration of performance properties of flat heating element.

Minor protrusions and dimples may be observed on the outer surface of the SC 90–1, SC 88 and SC 100-1 sealants surface layers over the whole application area.

Such blemishes as reeds (pores), gas bubbles or remains of incompletely melted powder were not observed. The thickness of surface structures ranges from 80 to 105 µm.



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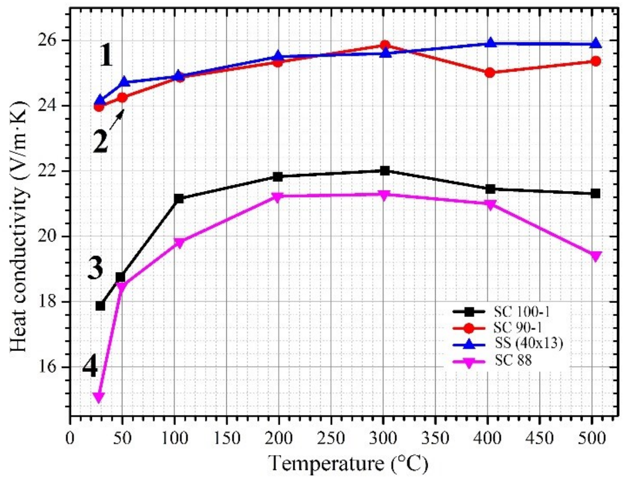

b

Fig. 6. Thermal diffusivity (a) and heat conductivity (b) of dielectric layers SC 90–1 (2), SC 100–1 (3) and SC 88 (4) on substrate of 40X13 stainless steel (1)

Therefore, taking into account comparatively low price of initial materials, high adhesion and electro-physical characteristics [5,6] obtained after heat treatment optimization, optimized glass-ceramic sealant chemistries of PbO–ZnO–B_2O_3–Al_2O_3–SiO_2–BaO system may be recommended to use as insulating coatings of flat heating elements.

Conclusion

The synthesis of glass-ceramic sealant of powders of PbO–ZnO–B_2O_3–Al_2O_3–SiO_2–(BaO) system according to the optimized chemical composition has been carried out. The heat treatment regime of dielectric surface layers of flat heating elements on 40X13 steel substrates is optimized. The advantages and disadvantages of sealants surface layers in comparison with dielectric surface layers applied according to the method of magnetron sputtering are shown. With regards to the low price of input materials and simplicity in the manufacturing process of dielectric surface layers production on the basis of glass-ceramic sealants, the technology may be introduced into the manufacturing process of flat heating elements.

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