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Electrical insulation test of alumina coating fabricated by sol–gel method in molten PbLi pool

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ABSTRACT

Development of electrical insulation coatings, which insulate an induced electric current from electrical conducting walls, is a key technology for the research and development of self-cooled liquid metal blanket including lead–lithium (PbLi) fusion blankets. As for magnetohydrodynamic (MHD) thermofluid study, an electrical insulation coating is extremely important from the viewpoint of the secure electrical insulating wall condition. The present study employs a sol–gel (SG) method to fabricate an Al_2O_3 coating, and discusses the feasibility of the SG coating as an electrical insulation coating for the PbLi through the electrical insulation test in the molten PbLi pool and the SEM with EDS analysis on the SG coating structure. The present study shows that the SG coating will be a potential electrical insulation coating for PbLi with both the operation time and the temperature limitation.

1. Introduction

A liquid metal flow in a strong magnetic field **B** induces an electric current **J**, and then interacts with **B**. As a result, the Lorentz force, $\mathbf{J} \times \mathbf{B}$ dominates the liquid metal flow under the strong magnetic field. The heat and mass transfers of the liquid metal flow under the strong magnetic field are also significantly affected by the Lorentz force, known as a magnetohydrodynamic (MHD) effect. The MHD thermofluid regime is determined by not only hydrodynamic and thermal conditions but also electric boundary conditions. A MHD duct flow confined by electrical conducting walls experiences a significant increase in pressure drop, known as MHD pressure drop.

Development of electrical insulation coatings, which insulate an induced electric current from electrical conducting walls, is a key technology for the research and development of self-cooled liquid metal blanket including lead–lithium (PbLi) fusion blankets [1]. As for MHD thermofluid study, an electrical insulation coating is extremely important from the viewpoint of the secure electrical insulating wall condition.

The dual-functional lead–lithium (DFLL) concept adopts two optional concepts of PbLi blankets including the reduced activation ferritic/martensitic (RAFM) steel-structured Helium-cooled quasi-static PbLi tritium breeder (SLL) blanket and the RAFM steel-structured Helium-gas/PbLi dual-cooled (DLL) blanket. DFLL concept considers an alumina (Al_2O_3) as a candidate for the electrical insulation coating against PbLi flows for the DLL blanket. The DFLL blanket concept employs a hot dip aluminizing (HDA) process, chemical vapor deposition (CVD) [2], or atmospheric plasma spray (APS) [3] to fabricate Al_2O_3 coatings on the RAFM steels. There are other purposes of coatings in fusion reactor blankets, depending on fusion blanket concepts. The water-cooled PbLi breeder (WCLL) concept with ferritic steel structures [4] adopts an Al_2O_3 coating as tritium permeation barrier (TPB) in order to reduce the tritium permeation into the water coolant caused by relatively high tritium partial pressure produced in the PbLi breeder to an acceptable level. In addition to the TPB, Al_2O_3 coatings have been studied as PbLi corrosion barrier because the PbLi solubility of Fe, Cr and particularly Ni are much higher than molten Li, resulting in greater dissolution than Li [5,6].

Most efforts of the Al_2O_3 coating development focus on fabricating an Al_2O_3 coating on ferritic/martensitic steels by means of HDA process [7], CVD [8], vacuum plasma spray (VPS) [9], low pressure plasma spray (LPPS) and APS [10]. The above investigations are originally not oriented for the Al_2O_3 electrical insulation coating against PbLi flows, but for the coating as TPB and corrosion barrier. Nevertheless, they are informative for the Al_2O_3 coating as electrical insulation coating in terms of the Al_2O_3 coating compatibility with the molten PbLi [11,12].

There are considerable requirements that must be satisfied in the coating for the fusion application. General requirements of coatings used for all fusion system are summarized as follows [1]:

(1) Potential for coating large complex geometry or configuration,

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(2) potential for in situ self-healing of defects that might occur,
(3) processing parameters should be compatible with material and capabilities, e.g. temperatures and processing durations,
(4) bonding/thermal expansion should match with substrate,
(5) acceptable neutronic properties,
(6) material availability/cost,
(7) safety/environmental characteristics, and
(8) radiation damage resistance.

In addition, electrical insulation coatings are required to have sufficient insulation efficiency. An electrical insulation efficiency of coatings is generally evaluated by the product of coating electric resistivity ρ_i and coating thickness δ_i, called as coating resistance $\rho_i\delta_i$. A previous study has estimated 10 μm thickness of an Al_2O_3, which has an electric resistivity $\rho_i \sim 10^{10}\ \Omega$ m at 400 °C, yields the almost perfect electrical insulation with $\rho_i\delta_i = 10^5\ \Omega\,m^2$ [13]. For the MHD thermofluid studies, some coating requirements may be eased because the thermofluid studies require a secure electrical insulation wall condition without neutron irradiation just for the experiment time period, which is much shorter compared to a time period of the fusion reactor operation.

The Al_2O_3 coatings fabricated on RAFM steels by HDA process and VPS have the favorable adhesion to its substrate even after being exposed in molten PbLi. However, the fabrication techniques are, in general, high cost and it is hard to fabricate the coatings on a large complex configuration. The present study employs a sol–gel (SG) method to fabricate an Al_2O_3 coating. The SG method is a wet-chemical technique used for fabricating ceramic materials, and it has the following advantages:

(1) Easy material availability,
(2) easy process to fabricate the Al_2O_3 coating on a substrate,
(3) applicable to large complex configurations, and
(4) low cost fabricating process.

On the other hand, the SG coating has poor adhesion to its substrate compared to the other processes such as HDA process.

The present study focuses on the Al_2O_3 coatings fabricated by SG method and discusses the feasibility that the SG coating works as electrical insulation coating for the purpose of PbLi MHD flow experiments, where the PbLi operation temperature is relatively lower, the PbLi exposure time is shorter compared to the fusion blanket operation, and the neutron irradiation is not applied.

2. Al_2O_3 coating by sol–gel method

The present study employs a commercial SG coating material: Ceramacoat™ 503-VFG-C (The Armco Products, Inc). The Ceramacoat™ is a single-component, Al_2O_3-filled, high-temperature (maximum durable temperature: 1650 °C), and electrical insulation coating material. The Ceramacoat™ contains Al_2O_3 and mono aluminum phosphate ($AlPO_4$) suspended in an inorganic binder system. The cured Al_2O_3 coating is $10^7\ \Omega$ m at room temperature in volume resistivity.

2.1. Coating fabrication

The SG coating hardly gets enough adhesion to a smooth-surface substrate after curing. Substrate surfaces are roughened and cleaned using a surface cleaner in order to remove mechanical oil leftovers before applying the Al_2O_3 coating material. Then, the Al_2O_3 coating material is applied to the surface-prepared substrates in a thin coat using a brush to maintain a uniform material thickness. The coating applied substrates are dried in the air for about 4 h at room temperature, then cured at 95 °C, 260 °C and 370 °C for 2 h at each temperature sing electric furnace. In the curing process, a rapid increase temperature of the coatings may cause a rapid evaporation of the coating binder. As results, some coating cracks and peelings might occur. Therefore we employed the step curing to avoid the effect by following the coating company instructions.

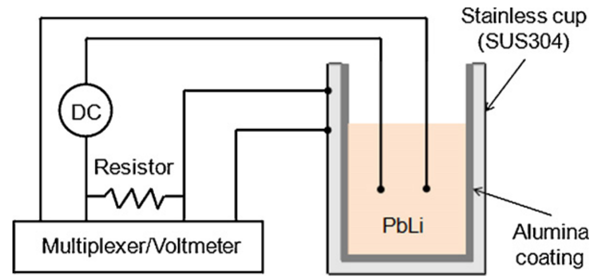

Fig. 1. Electrical insulation test.

3. Electrical insulating test of the Al_2O_3 coating in a PbLi pool

There are considerable studies performed on coating issues. However, attentions have been paid mainly to coating fabrication techniques, the tritium permeation and corrosion behaviors through morphology of the corrosion layers and discussions on chemical reactions between the liquid metal and wall material [11,12,14].

There is no research on electric insulation performance of the coatings in contact with liquid metal. The SG coating is expected to be less durable against thermal expansion differences because the SG coating has poor adhesion. An electrical insulation test of the SG coating in a PbLi pool was carried out to discuss the electrical insulation performance in contact with the molten PbLi, and then discuss the feasibility of the SG coating as electrical insulation coating for MHD thermofluid research.

3.1. Experimental setup

Electrical insulating tests in a molten PbLi pool were performed to examine an electrical insulating performance of the Al_2O_3 coating fabricated by the SG method. The SG coatings were fabricated on SUS304 cups used in the test by going through the above-mentioned procedure. Fig. 1 shows the setup of the electrical insulating test. The Al_2O_3-coated cups were filled with the molten PbLi in an electric furnace. One side of electrodes was attached to the cup substrate, and the other side of electrodes was immersed in the molten PbLi. The electrodes were connected to a power supply to apply a few volts on them. They were also connected to a multiplexer to measure an electric current passing through the SG coating with time variation. The PbLi temperature, the air temperature in the electric furnace and the electric current passing through the SG coating were measured every 10 min by the multiplexer. The following two kinds of test runs were conducted;

Run #1: molten PbLi temperature was at 300 °C for around 170 h. (Isothermal)
Run #2: molten PbLi temperature was increased from 300 °C up to 500 °C. (Heating-up test)

In a design of PbLi blanket, it is proposed to operate at higher temperature. However, the present study is primarily oriented for the PbLi MHD thermofluid. The experiment temperatures were determined from a viewpoint of keeping the PbLi melt during experiments. The PbLi melting temperature is 235 °C. The present temperature range (around 300–500 °C) is high enough for the PbLi

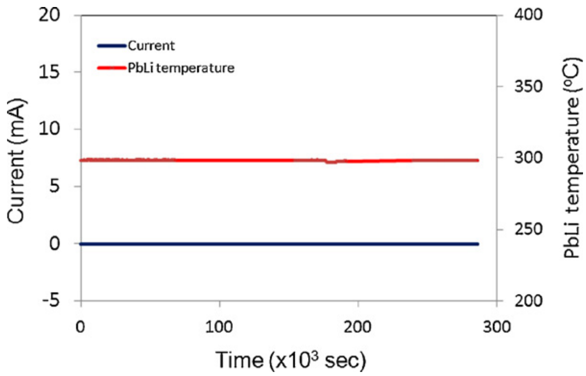

Fig. 2. Electrical insulation test result (Run #1).

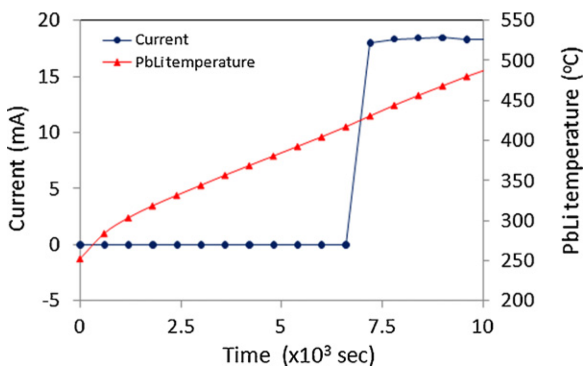

Fig. 3. Electrical insulation test result (Run #2).

MHD thermofluid because similarity laws exist in heat transfer and flows, and eliminates some problems related to a high-temperature operation, such as an enhancement of the PbLi corrosion.

The PbLi eutectic alloy was prepared by the Atlantic Metals & Alloys, Inc. The chemical composition of the PbLi alloy was 81 at% in Pb and 19 at% in Li with some impurities (Cu, Zn, Fe, Ag, Bi and Si).

3.2. Results and discussion

The results of electrical insulation test are shown in Figs. 2 and 3. Fig. 2 shows the results of Run #1, indicating that the SG coating insulated the electric current between the molten PbLi at 300 °C and its substrate for around 170 h. Fig. 3 shows the results of Run #2, indicating that the electrical insulation coating was broken to allow the electric current at around 430 °C. Another similar test was conducted, and showed the electrical insulation was broken

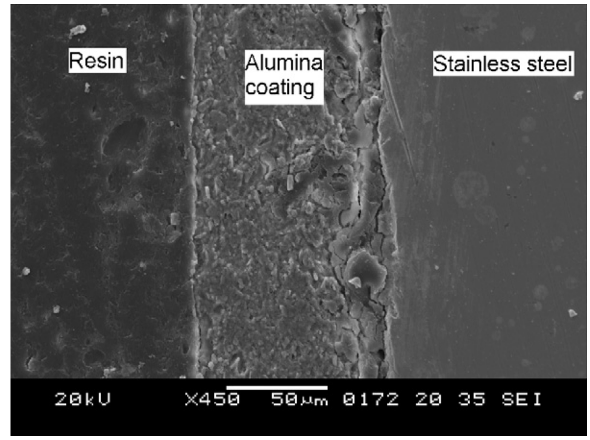

Fig. 5. SEM image of the Al_2O_3 coating cross-section without experience of molten PbLi exposure during the electrical insulating test.

at around 410 °C. These results indicate the SG coating cured up to around 370 °C have a threshold for the electrical insulation breaks above 400 °C of the molten PbLi temperature.

4. SEM observation with EDS analysis

4.1. Results and discussion

Scanning electron microscopy (SEM) observations together with energy-dispersive X-ray spectroscopy (EDS) were conducted to examine the cured SG coating surface, structure and thickness. The SG coating structure is significant for the coating compatibility with the molten PbLi. Fig. 4 shows the SEM images of the SG coating surface. The SEM images revealed that the SG coating consisted of integrated network of around 5 µm particles.

In order to observe cross-sections of the SG coating after the electrical insulation test, the SG coated cup was cut into some pieces before a resin impregnation, and consecutively a surface polishing process. Figs. 5 and 6 show the SEM images of SG coating cross-sections exposed and unexposed to the molten PbLi in Run #1 of the electrical insulation test, respectively.

In Fig. 5, a coarser area in between the bulk SG coating and substrate was the roughened substrate surface. It depicted that the SG coating was combined to its roughened substrates not chemically but physically. That is why the SG coating adhesion is weaker than the other processes such as the HDA process. The SEM observation also showed the SG coatings were around 100 µm of average thickness with concave–convex surfaces. Based on these results, it was found that the SG coating was approximately 10 times more effec-

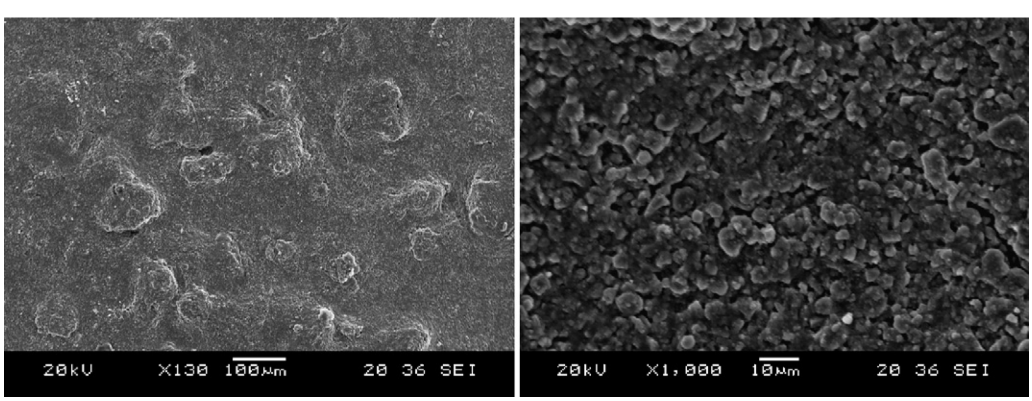

Fig. 4. SEM images of the Al_2O_3 coating surface (left: ×130, right: ×1000).

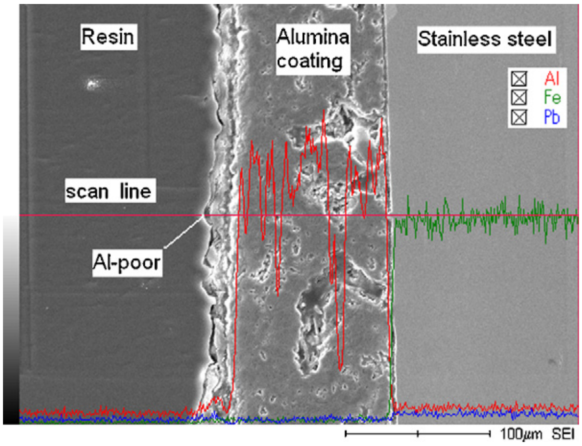

Fig. 6. SEM image of the Al_2O_3 coating cross-section with experience of molten PbLi exposure during the electrical insulating test.

tive in the coating resistance than the almost electrical insulation value: $\rho_i\delta_i = 10^5\ \Omega\,m^2$, assuming that the SG coating has same electrical resistivity as employed in the coating resistivity estimation [13].

Fig. 6 showed the SG coating surface was corroded by the molten PbLi by the depth of around 15–25 μm, and Al in the corroded SG coating layer decreased after the 170 h exposure in the molten PbLi, however Pb in the Al-poor layer was still small. Therefore, it was concluded that Al in the SG coating was transferred into the molten PbLi, but the SG layer did not allow the molten PbLi to permeate into itself in the present study. If the SG coating is exposed in the molten PbLi flows, the PbLi corrosion will be more serious than in the present study. More experiments on the corrosion behavior in the PbLi flows are necessary to examine whether or not the corrosion rate is within an acceptable level.

Fig. 6 also depicted that there were large pores in the SG coating near its substrate. The large pore will increase the entire electrical resistivity of the SG coating. On the other hand, they may enhance the coating peelings or cracks.

4.2. Discussion on the electrical insulation break

A mismatch of thermal expansion is a dominant parameter for coating adhesion durability. In the present study, a thermal expansion mismatch between the Al_2O_3 coating and its substrate caused the coating detachment or the crack with the PbLi temperature above the curing temperature, resulting in the electrical insulation breaks. A previous study [15] showed that an undercoating between a stainless steel and an Al_2O_3 coating with the coefficient of thermal expansion close to Al_2O_3 could overcome the coating cracks or peelings. In addition to the mismatch of thermal expansions, poor adhesion of the Al_2O_3 coating caused coating cracks or peelings more easily. The SEM together with EDS analysis showed that the PbLi corrosion did not reach the substrate, only in the surface region of the SG coating. Therefore, all the results indicate that the mismatch of the thermal expansions is the primary contributor to the electrical insulation break.

The curing processes and conditions are also dominant parameters for the SG method to determine the coating structure. Curing the SG coatings at higher temperature may improve the SG coating performance to be durable up to the higher curing temperature. More experiments on the curing temperature issue are necessary for the detailed discussion.

5. Conclusions

The present study discusses the feasibility of the Al_2O_3 coating fabricated by the SG method as an electrical insulation coating for molten PbLi through the electrical insulation test in the molten PbLi pool and the SEM together with EDS analysis on the SG coating structure. The present study conclusions are summarized as follows:

(1) The SG coatings worked as an electrical insulating coating for 170 h in a molten PbLi pool at 300 °C in the present study. The SG coating performance in longer operation still remains to be evaluated. Nevertheless, the SG coating fabricated will have a potential electrical insulation for PbLi MHD thermofluid studies with both the operation time and the temperature limitation.
(2) The present results indicate the SG coating cured up to around 370 °C have a threshold for the electrical insulation break above 400 °C. More experiments on the curing temperature issue are necessary for the detailed discussion.
(3) The SEM with EDS analysis revealed the structural characteristics of the SG coating, and the PbLi corrosion occurred during the electrical insulation test, indicating that the primary contribution to the electrical insulation break was not from the PbLi corrosion, but was due to the mismatch of the thermal expansions between the SG coating and the substrate.

Further investigations on material interactions and chemistry control issues associated with the fabrication process, the stability and the performance of the SG coating are necessary even for the usage in PbLi MHD thermofluid study.

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