# Consideration of Alumina Coating Fabricated by Sol-gel Method as MHD Coating against PbLi

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### ABSTRACT

Development of effective electrical magnetohydrodynamic (MHD) coating insulating induced current path is one of key fusion technologies for fusion blanket research and development. Electrical insulation coating is extremely important for lead lithium (PbLi) MHD thermofluid study from pointview of secure electric insulating wall condition. The alumina (Al<sub>2</sub>O<sub>3</sub>) coatings fabricated by a sol-gel method have never studied as electrical insulation coating against PbLi as a working fluid. The present paper discusses the feasibility of the Al<sub>2</sub>O<sub>3</sub> coating by the sol-gel method as an electrical insulation coating for PbLi MHD flows. It is examined the electrical insulation durability of the Al<sub>2</sub>O<sub>3</sub> coating by the sol-gel method with molten PbLi exposed. The Al<sub>2</sub>O<sub>3</sub> coatings worked as electrical insulation coating for 170 hours with PbLi temperature at 300 °C in this study. However, the Al<sub>2</sub>O<sub>3</sub> coating cured up to around 370 °C seemed to have a threshold for the electric insulation break above 400 °C. Nevertheless, the Al<sub>2</sub>O<sub>3</sub> coating fabricated by the sol-gel method will be a potential electric insulation coating for PbLi flow under the MHD condition with the operation time and the temperature limitation.

### **KEYWORDS**

PbLi, MHD coating, alumina coating, sol-gel method

### 1. INTRODUCTION

In some liquid metal fusion blanket concepts, lead-lithium eutectic alloy (PbLi) circulates as a breeder (e.g. helium-cooled lead lithium (HCLL) blanket concept [1]), or both as a breeder and a coolant (e.g. dual-coolant lead lithium (DCLL) blanket concept [2]). The liquid metal motion in a strong magnetic field **B** induces an electric current **J**, and then interacts with the

strong magnetic field. As a result, the Lorentz force, **JxB** dominates the liquid metal flow under the strong magnetic field. The heat and mass transfer of the liquid metal flow under a strong magnetic field are also significantly affected by the Lorentz force, known as magneto-hydro-dynamic (MHD) effects. MHD thermofluid regime is determined by not only hydrodynamic and thermal conditions but also eletctric boundary conditions. MHD duct flow confined by electric conductive walls experiences significant increase in pressure drop known as MHD pressure drop.

Development of effective and reliable coatings is one of key fusion technologies for fusion blanket research and some fusion blanket concepts themselves [3]. Most of the efforts on electrical insulation coating development have been made for the self-cooled lithium concepts with a vanadium alloy structure (Li/V) [4-6]. As for PbLi electrical insulation coating, dual functional lead lithium (DFLL) adapts two optional concepts of PbLi blankets including the reduced activation ferritic/martensitic (RAFM) steel-structured He-cooled quasi-static PbLi tritium breeder (SLL) blanket and the RAFM steel structured He-gas/ PbLi dual-cooled (DLL) blanket. DFLL concept considers an alumina (Al<sub>2</sub>O<sub>3</sub>) as a candidate for the electric insulation coating against PbLi flow for the DLL blanket [7]. The Al<sub>2</sub>O<sub>3</sub> coating for DFLL blanket is fabricated on a RAFM steel by means of a hot dip aluminizing (HDA) and a chemical vapor deposition (CVD), and the Al<sub>2</sub>O<sub>3</sub> coating compatibility with molten PbLi is examined. There is another technique to minimize MHD pressure drop as an alternative technique for electric insulation coatings, DCLL blanket design adapts SiC<sub>4</sub>/SiC composite or metallic flow channel insert (FCI) for thermal and electric insulators both to keep its RAFM steel structure below acceptable temperature and reduce significant MHD pressure drop.

There is another purpose of coatings in fusion blankets, depending on fusion blanket concepts. The water-cooled PbLi breeder (WCLL) concept with ferritic steel structures [8] adapts a coating for tritium permeation barrier (TPB) in order to reduce tritium permeation into water coolant caused by relatively high tritium partial pressure produced in PbLi breeder to an acceptable level. Most of the efforts on the TPB coating development are focused on the Al<sub>2</sub>O<sub>3</sub> coatings fabricated on aluminized ferritic/martensitic steels by means of HDA [9], CVD [10], a vacuum plasma spray (VPS) [11], a detonation jet, a low pressure plasma spray (LPPS) and an air plasma spray (APS) [12]. These investigations are originally not oriented for electric insulation the Al<sub>2</sub>O<sub>3</sub> coating against the molten PbLi, but for the coating as TPB. Nevertheless, the investigations are also useful for the Al<sub>2</sub>O<sub>3</sub> coating as the electric insulation coating especially in terms of the coating compatibility with the molten PbLi.

There are considerable requirements that must be satisfied in the coating development for the fusion application. General requirements of coatings used for all fusion system are summarized as follows [3]:

- 1) Potential for coating large complex geometry or configuration,
- 2) Potential for in site self-healing of defects that might occur,
- 3) Processing parameters compatible with material and capabilities, e.g. temperatures and times,
- 4) Bonding/thermal expansion match with substrate,
- 5) Acceptable neutronic properties,
- 6) Material availability/cost,
- 7) Safety/environmental characteristics, and
- 8) Radiation damage resistance.

In addition to the requirements, electric insulation coatings must have sufficient insulation efficiency to electrically decouple a wall substrate and a working fluid. The electric insulation coating performance is generally evaluated by the product of coating electric resistivity  $\rho_i$  and

coating thickness  $\delta_i$ , called as coating resistance  $\rho_i \delta_i$ . A previous estimation [13] reported 10  $\mu$ m thickness Al<sub>2</sub>O<sub>3</sub> coating at 400 °C provides almost perfect electric insulation with  $\rho_i \delta_i = 10^5 \Omega m^2$ . Electric insulation coatings are desired to satisfy all the requirements mentioned above.

The  $Al_2O_3$  coatings fabricated on aluminized ferritic/martensitic steels by means of HDA and CVD are reported to have the favorable bonding with its substrate against the molten PbLi exposed. However, the fabrication techniques are, in general, high cost and difficult to fabricate on a large complex configuration. The sol-gel method of  $Al_2O_3$  coating has some advantages as follows:

- 1) Easy material availability,
- 2) Easy process to fabricate Al<sub>2</sub>O<sub>3</sub> coating on a substrate,
- 3) Applicable to large complex configuration, and
- 4) Low cost fabricating process.

On the contrary, the  $Al_2O_3$  coating fabricated by the sol-gel method has poor bonding to its substrate compared to the above-mentioned process e.g. HDA and CVD.

The  $Al_2O_3$  coatings fabricated by the sol-gel method have never been studied as the electrical insulation coating against PbLi flows. The present research focuses on the  $Al_2O_3$  coatings fabricated by the sol-gel method and discusses the feasibility of the  $Al_2O_3$  coating as the electrical insulation coating for PbLi flows under the MHD condition.

### 2. Al<sub>2</sub>O<sub>3</sub> COATING BY SOL-GEL METHOD

The sol-gel method fabrication of  $Al_2O_3$  coating has been commercialized already. Aremco Products, Inc. commercially provides an  $Al_2O_3$  coating material (Ceramacoat<sup>TM</sup> 503-VFG-C), which can be fabricated by the sol-gel method. The Ceramacoat<sup>TM</sup> is a single-component,  $Al_2O_3$ -filled, high-temperature (maximum durable temperature: 1650 °C), and electrical insulation coating material. The  $Al_2O_3$  coating material contains an aluminum oxide and a mono aluminum phonosphate suspended in an inorganic binder system. The cured  $Al_2O_3$ coating is  $10^7 \Omega$  m at room temperature in volume resistivity and 253 V/mm at room temperature in dielectric strength.

#### 2.1. Fabrication

Substrate surfaces are cleaned thoroughly prior to application of the  $Al_2O_3$  coating material. Extremely smooth substrate surfaces are hard to bond. Substrate surfaces are roughened and cleaned using a surface cleaner in order to remove mechanical oils remaining on the surface as surface preparation before application of 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 of 2-8 mm. The substrates with the  $Al_2O_3$  coating material applied are air set for 1-4 hours at room temperature, then step cured at 93 °C, 260 °C and 372 °C for 1-2 hours at each temperature using electric furnace.

The  $Al_2O_3$  coating has poor electrical insulation in sharp configuration such as corner or edge. That is because the  $Al_2O_3$  coating stuff hardly covers sharpened substrates even before curing process. Rounded substrates or flat plates are easily covered with the  $Al_2O_3$  coating stuff. Fig. 1 shows the picture of  $Al_2O_3$  coating fabricated inside a stainless steel cup.

### 2.2. SEM observation

Scanning electron microscopic (SEM) observation was conducted to examine the cured  $Al_2O_3$  coating surface and structure on its substrates. The  $Al_2O_3$  coating structure is a significant parameter for the coating durability and the compatibility with molten PbLi. Fig. 2 and 3 show the SEM images of the  $Al_2O_3$  coating surface. They show the  $Al_2O_3$  coating consists of around 5 µm particles. The  $Al_2O_3$  coating thickness is also a significant parameter for electrical insulation efficiency shown above. Material analysis of the  $Al_2O_3$  coating cross-section is future work.



Fig. 1 The Al<sub>2</sub>O<sub>3</sub> Coating Fabricated inside a Round Cup



Fig. 2 SEM Image of the Al<sub>2</sub>O<sub>3</sub> Coating Surface (x130)



Fig. 3 SEM Image of the Al<sub>2</sub>O<sub>3</sub> Coating Surface (x1000)

### 3. ELECTRICAL INSULATION TEST OF Al<sub>2</sub>O<sub>3</sub> COATING

With the molten PbLi exposed, the electrical insulation performance of the Al<sub>2</sub>O<sub>3</sub> coating fabricated by the sol-gel method was examined.

### 3.1. Experimental set up

The  $Al_2O_3$  coating is fabricated by the sol-gel method on stainless steel cups in the procedure mentioned above. The stainless cup is made of SUS304. Fig. 4 and 5 show the  $Al_2O_3$  coating electrical insulation test set-up. The stainless cups with the  $Al_2O_3$  coating were filled with the molten PbLi in an electric furnace. Electrodes were attached to one for the cup substrate, and the other for immersing into the molten PbLi. The electrodes were connected to a power supply to be applied a few volts, and to a Multiplexer/Voltmeter to measure the current passing through the  $Al_2O_3$  coating with time variation. The PbLi temperature, air temperature in the electric furnace and the current passing through the  $Al_2O_3$  coating were sampled every 10 minutes by the multiplexer. The following 3 test runs were conducted with the change of PbLi temperature as the experimental parameter;

Run #1: PbLi temperature was controlled at 300 °C exposing for around 170 hours. Run #2: PbLi temperature was controlled to increase up to 500 °C. Run #3: same as Run #2.

#### 3.2. PbLi alloy

The PbLi alloy used in the test is prepared to be (83at%Pb-17at%Li) eutectic alloy. The alloy contains Pb and Li as bulk chemical composition and several elements as impurity substance shown in Table 1.



# Fig. 4 Electrical Insulation Test



Fig. 5 Electrical Insulation Test

Table 1. PbLi chemical composition analysis from the vender(Atlantic Metals & Alloys, Inc.)

	Wt %	at %	ppm
Pb	99.19	80.92	-
Li	0.78	18.99	-
Ag	0.0010	0.0016	15.89
Bi	0.0100	0.0081	80.88
Cu	0.0010	0.0027	26.60
Fe	0.0050	0.0151	151.33
Si	0.0100	0.0602	601.81
Zn	0.0010	0.0026	25.85

#### 3.3. Results and discussion

The electrical insulation test results are shown in Figs. 6-8. Fig. 6 shows the results of Run #1. The  $Al_2O_3$  coating insulated current path between the molten PbLi and its substrate with exposed to the molten PbLi at 300 °C for around 170 hours. Fig. 7 shows the results of Run #2. With increase in temperature, the electrical insulation of the  $Al_2O_3$  coating was broken to let the current pass through the coating at around 430 °C of PbLi. Fig. 8 shows the results of Run #3. It indicates the similar result to Run #2. With increase in temperature, the electrical insulation of the  $Al_2O_3$  coating was broken around 410 °C in PbLi.

Run #1 result indicated the  $Al_2O_3$  coating fabricated by the sol-gel method has the electrical insulating durability for approximately 170 hours in this study. Run #2 and #3 results indicated the  $Al_2O_3$  coating cured up to around 370 °C seemed to have a threshold for the electric insulation break above 400 °C.

The coefficients of thermal expansion of Stainless steel 304 and alumina are  $17.3 \times 10^{-6}$  and around  $8.0 \times 10^{-6}$  1/ °C, respectively. The thermal expansion mismatch between the Al<sub>2</sub>O<sub>3</sub> 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. The previous study [14] shows the undercoating between a stainless steel and alumina coating with the coefficient of thermal close to alumina can overcome the coating cracks or peelings. The mismatch of thermal expansion is considered to be a dominant parameter in terms of the coating durability. The alumina coatings by the sol-gel method combine to their substrates not chemically, but physically. In addition to the mismatch of thermal expansion, the coating poor bonding caused the coating cracked or peeled easier compared to the other methods e.g. HDA. The curing process and temperature are also dominant parameters for the coating structure or durability by the sol-gel method. The Al<sub>2</sub>O<sub>3</sub> coating with cured at higher temperature will have a potential as the effective electrical insulation coating endurable up to the higher temperature.



Fig.6 Electrical Insulation Test Result (Run #1)



Fig. 7 Electrical Insulation Test Result (Run #2)



Fig. 8 Electrical Insulation Test Result (Run #3)

More experiments on the curing temperature issue must be necessary for the detailed discussion of the  $Al_2O_3$  coating performance. The present results also indicted the  $Al_2O_3$  coatings worked as electrical insulation coating for 170 hours with the PbLi temperature at 300 °C. However, the  $Al_2O_3$  coating performance in longer operation still remains to be evaluated.

In addition, material interaction and chemistry control issues associated with fabrication process, stability and performance of the  $Al_2O_3$  coating should be discussed for the usage in fusion blanket, and even in PbLi MHD thermofluid studies.

# 4. CONCLUSIONS

The present paper describes the  $Al_2O_3$  coatings by the sol-gel method, and discusses the feasibility of the  $Al_2O_3$  coating as an electrical insulation coating for PbLi flow under the MHD condition. The electrical insulation durability of the  $Al_2O_3$  coating by the sol-gel method with molten PbLi exposed is examined.

The present research conclusions are summarized as follows:

- The Al<sub>2</sub>O<sub>3</sub> coatings worked as electrical insulating coating for 170 hours with PbLi temperature at 300 °C in this study. However, the Al<sub>2</sub>O<sub>3</sub> coatings performance in longer operation still remains to be evaluated. Nevertheless, the Al<sub>2</sub>O<sub>3</sub> coating fabricated by the sol-gel method will have a potential electric insulation for PbLi MHD thermofluid studies with the operation time and the temperature limitation.
- 2) The Al<sub>2</sub>O<sub>3</sub> coating cured up to around 370 °C seemed to have a threshold for the electric insulation break above 400 °C. It is discussed that the Al<sub>2</sub>O<sub>3</sub> coating with cured at higher temperature will have a potential as the effective electrical insulation coating endurable up to the higher temperature.

Further investigations on the  $Al_2O_3$  coating must be necessary for the detailed discussion of the coating performance. The insulation coating thickness is a significant parameter for electrical insulation efficiency. Material analysis of the  $Al_2O_3$  coating cross-section must be conducted in order to evaluate the electrical insulating efficiency. In addition, the curing process and temperature are dominant parameters for the sol-gel method coating structure or durability. From the viewpoint of evaluation of the coating durability, the material analysis of the  $Al_2O_3$  coating cross-section must be necessary. Material interaction and chemistry control issues associated with the fabrication process, the stability and the performance of the  $Al_2O_3$  coating should be discussed for the usage in fusion blanket, and even in PbLi MHD thermofluid studies.

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# REFERENCES

- 1. G. Rampal, et al. "HCLL TBM for ITER-Design Studies, *Fusion Eng. Des.*, **75-79**, 917-922, (2005)
- 2. C. P. C. Wong, et al., "An Overview of Dual Coolant Pb-17Li Breeder First Wall and Blanket Concept Development for the US ITER-TBM Design", *Fusion. Eng. Des.*, **81**, 461-467, (2006)
- D. L. Smith, J.-H. Park, I. Lyublinski, V. Evtikhin, A. Perujo, H. Glassbrenner, T. Terai, S. Zinkle, "Progress in Coating Development for Fusion Systems" *Fusion Eng. Des.*, 61-62, 629-641, (2002)
- 4. S. Malang, H.U. Borgstedt, E.H. Farnum, K. Natesan, I.V. Vitkovski, "Crucial Issues on Liquid Metal Blanket Design", *Fusion Eng. Des.*, **16**, 95–109 (1991)
- D.L. Smith, K. Natesan, J.-H. Park, C.B. Reed, R.F.Mattas, "Development of Electrically Insulating Coatings on Vanadium Alloys for Lithium-cooled Blankets", *Fusion Eng. Des.*, 51–52, 185–192 (2000)
- T. Muroga, J.M. Chen, V.M. Chernov, K. Fukumoto, D.T. Hoelzer, R.J. Kurtz, T.Nagasaka, B.A. Pint, M. Satou, A. Suzuki, H. Watanabe, "Review of Advances in Development of Vanadium Alloys and MHD Insulator Coatings", *J. Nucl. Mater.*, 367– 370, 780–787 (2007)
- 7. Y. Wu, et al, "Conceptual Design Activities of FDS Series Fusion Power Plants in China", *Fusion Eng. Des.*, **81**, 2713-2718, (2006)
- L. Giancarli, L. Baraer, B. Bielak, M. Eid, M. Futterer, E. Proust, X. Paepsaet, J.F. Salavy, L. Sedano. "European Reference Design of the Water-cooled Lithium-lead Blanket for a Demonstration Reactor", *Fusion Technology*, 26, 1079-1085, (1994)
- 9. H. Glassbrenner, K. Stein-Feechner, J. Konys. "Scale Structure of Aluminzed Manet Steel after HIP Treatment", *J. Nucl. Mater.*, **283-287**, 1302-1305, (2000)
- 10. C. Chabrol, E. Rigal, F. Schuster, Report CEA, No. 73/97, CEA Grenoble, (1997)
- A. Perujo, T. Sample, E. Serra, H. Kolbe, "Low Aluminum Content Permeation Barrier Coatings for din 1.4914 Martensitic Steel (Manet)", *Fusion Technology*, 28, 1256-1261, (1995)
- 12. G. Benamati, C. Chabrol, A. Perujo, E. Rigal, H. Glassbrenner, "Development of Tritium Permeation Barriers on Al Base in Europe", *J. Nucl. Mater.*, **271-272**, 391-395, (1999)
- 13. L. Buhler, S. Molokov, "Magnetohydrodynamic Flow in Ducts with Insulating Coatings" *Report KfK 5103*, Forschungzentrum Karlsruhe, (1993)
- 14. M.Nakamichi, H. Kawamura, "Out-of-pile Characterization of Al<sub>2</sub>O<sub>3</sub> Coating as Electrical Insulator", *Fusion Eng. Des.*, **58-59**, 719–723 (2001)