Research Article

Simulation of the Residual Stress of the Y_2O_3/Al_2O_3 Composite Deuterium Permeation Barrier under Thermal Shock

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A deuterium permeation barrier is an essential part in the core component of nuclear reactors. It can protect the structure made of steel from being penetrated by deuterium in a fusion reactor. However, residual stress induced in the operation would dramatically influence the mechanical endurance of the coating, threatening the safety of the facilities. In this paper, finite element analysis was conducted to investigate the residual stress in nanoscale Al_2O_3 and Y_2O_3 coatings and their composites under thermal shock, from 700°C to 25°C. The max principal stress is assumed as the cause of crack initiation in the coating, because ceramics are brittle and fragile under tensile stress. Max shear stress and max Mises stress in the systems are also analyzed, and the effect of thickness in the range 100 nm to 1000 nm was investigated. The max principal stress in Al_2O_3 coating reaches its maximum value, 1.33 GPa, when the thickness of coating reaches 450 nm. And the max principal stress decreases at a very low rate as the thickness increases exceeding 450 nm. The max principal stress in Y_2O_3 coating increases rapidly as the thickness increases when the thickness of the coating is below 250 nm, and the max principal stress is at about 0.9 GPa when the thickness exceeds 500 nm. The max principal stress in the Y_2O_3/Al_2O_3 (150 nm) composite coating occurs in the Al_2O_3 layer and shows no difference from the single layer of 150 nm thick Al_2O_3 coating. The max principal stress site of all three kinds of coating is located at the edge of the coating 25 nm away from the interface. The result shows that residual thermal stress in the coating increases as the thickness increases when the thickness of the coating is below 200 nm due to the stress singularity of the interface. And as the thickness exceeds 500 nm, the increase in thickness has little impact on the residual thermal stress in the coating. Coating an Y_2O_3 top layer will not introduce any more residual thermal stress under the thermal shock condition. The Y_2O_3 coating causes much less residual stress under thermal shock compared with Al_2O_3 owing to its much lower Young's modulus. The max principal stress in the 300 nm thick Y_2O_3 coating is 0.85 GPa while that of the Al_2O_3 coating is 1.16 GPa. The max residual stress of the composite Y_2O_3/Al_2O_3 (150 nm) coating is determined by the Al_2O_3 layer.

1. Introduction

Deuterium permeation is one of the most critical threats to the safety of fusion reactors. Deuterium has strong reducibility and ability to be dispersed in other materials for most structural materials. When deuterium is dispersed into the structural material, it can cause nuclear fuel leakage, contam-

ination, and material embrittlement which can lead to structure failure [1–3]. In order to solve this critical problem, ceramic coatings, including the oxides (Al_2O_3 , Cr_2O_3 , Y_2O_3 , and Er_2O_3), the nitrides (Fe_3N , TiN), the carbides (SiC , TiC), and their composites, are applied to act as a deuterium permeation barrier [4–7]. Among all kinds of ceramic coatings, oxide ones are the most common choice because of their

high deuterium permeation resistance and low cost. Al_2O_3 has been proven to be ideal coating to prevent deuterium gas-driven permeation. This coating could significantly reduce deuterium penetration into the substrate. The combination of Al_2O_3 coating and 316L stainless steel substrate, with high strength, high deuterium permeation resistance, strong thermodynamic stability, and relatively low cost, is the ideal candidate to construct fusion reactors [8–11]. However, Al_2O_3 coating has a huge thermal mismatch with the substrate, 316L stainless steel, because their coefficient of thermal expansions differs widely, causing tremendous residual thermal stress after the system endures thermal shock during processing and serving, such as the operation of the fusion reactor, thus threatening the bond between the coating and the substrate, causing cracks initiating and propagating near the interface [12]. Many efforts have been made to analyze the residual thermal stress in the coating/substrate system in search of the failure mechanism and the optimization methods, most of which are on the micron dimension [13–19]. This paper advances further into nanodimension, because nanothick coatings show better mechanical performance [20, 21], investigating the effect of thickness on the residual stress of nanoscale composite $\text{Al}_2\text{O}_3/\text{Y}_2\text{O}_3$ coating under thermal shock via finite element analysis (FEA), as previous study reveals that $\text{Al}_2\text{O}_3/\text{Y}_2\text{O}_3$ composite coating has better deuterium permeation resistance than the single-layer Al_2O_3 coating due to the interface between the layers [21]. And the thermal residual stress distribution of the coatings was also analyzed to determine the most likely crack initiation site in the coatings. The research is the preliminary work for understanding the crack initiation and propagation mechanism of the nanoscale deuterium permeation barrier and the optimization of nanoscale permeation barrier preparation.

2. Material and Methods

ANSYS 19 is employed to simulate the stress field of the coatings under thermal shock. All models are considered stress-free under 700°C . Because specimens were annealed in a vacuum tube furnace for 2 h at 700°C , the residual stress produced during the deposition process should be eliminated [21]. Max principal stress and max shear stress and max Mises stress are analyzed. Max principal stress is the major criterion of the coating because oxide ceramics are considered brittle material and fragile under tensile stress.

2.1. Analytic Model. In the finite element analysis, displacements are the solution factors that are stored in nodal positions. Loads are defined as prescribed forces and displacements. Thus, the strain and stress increments at any point in the element can be calculated with the interpolation functions. ANSYS transforms those mechanical equilibrium equations into simultaneous equations. And the displacements and forces can be calculated via figuring out elemental stiffness matrixes.

The total strain vector, $\{\Delta\epsilon\}$, can be expressed as

$$\{\Delta\epsilon\} = \{\Delta\epsilon^{\text{el}}\} + \{\Delta\epsilon^{\text{th}}\}, \quad (1)$$

where $\{\Delta\epsilon^{\text{el}}\}$ is the elastic strain increment vector and $\{\Delta\epsilon^{\text{th}}\}$ is the thermal strain increment vector.

Within each element, the relation between $\{\Delta\epsilon\}$ and the strain matrix, $\{B\}$, fulfill the following equation:

$$\{\Delta\epsilon\} = \{B\}\{\delta\}^e, \quad (2)$$

where $\{\delta\}$ is the displacement vector for any given element (e).

The elastic stress increment vector, $\{\Delta\sigma^{\text{el}}\}$, can be calculated with Hooke's law:

$$\{\Delta\sigma^{\text{el}}\} = \{D\}\{\Delta\epsilon^{\text{el}}\}, \quad (3)$$

where $\{D\}$ is the elastic matrix related to the elastic modulus, E , and Poisson's ratio, ν , for the given material at given temperature.

The thermal stress increment vector, $\{\Delta\epsilon^{\text{th}}\}$, can be calculated as follows:

$$\{\Delta\epsilon^{\text{th}}\} = \{\alpha\}\Delta T, \quad (4)$$

where α is the thermal expansion coefficient and ΔT is the temperature increment.

Stresses are calculated by applying the principle of virtual work.

$$\{\delta\}^T \{\Delta F\}^e = \iiint \{\epsilon\}^T \{\sigma\} dx dy dz. \quad (5)$$

Substitution of equations (3) and (4) into (5) gives

$$\{\Delta F\}^e = \iiint \{B\}^T \{D\} \{\Delta\epsilon^e\} dx dy dz. \quad (6)$$

The element stiffness matrix $\{K\}$ fulfills the equilibrium equation:

$$\{K\}\{\delta\} = \{F\}. \quad (7)$$

In the thermal shock condition, the load vector $\{\Delta F\}$ only includes the thermal force. And the thermal stress can be derived as follows:

$$\{\Delta F\}_T^e = \iiint \{B\}^T \{D\} \{B\} \{\alpha\} dx dy dz. \quad (8)$$

2.2. Model Geometry and Material Properties. The models simulate coatings prepared on the 316L stainless steel by radio-frequency magnetron sputtering. The morphologies of the systems in SEM (Hitachi-S4800) are shown in Figure 1. The nanoscale coatings were dense and homogeneous according to the pictures. And the interface between layers and the surface of the top coating was rather smooth, and no obvious defects were observed before the thermal shock test. More detailed information about the preparation and the characterization of the coatings can be checked in [21] and are shown in Figure 2.

The model used in the analysis is made up of 2 parts. One is a cylinder-shape 316L stainless steel substrate with 1 mm in diameter and 0.5 mm in thickness. The other is a coating with

FIGURE 1: SEM images of the nanoscale coatings [21]: (a) Al_2O_3 ; (b) Y_2O_3 ; (c) $\text{Y}_2\text{O}_3/\text{Al}_2\text{O}_3/316\text{L SS}$.

thickness, t , deposited on the top surface of the 316L SS substrate. Each part is assumed to be uniform and homogenous. Thus, 2-dimensional axisymmetric models are applied to simplify the analysis. The properties of the materials are shown in Table 1.

2.3. Meshing. In order to construct a nanoscale model with an adequate space for meshing, the standard unit of length in all models is set to micron. Thus, all parameters that involve length are converted to match the change (e.g., Young's modulus of the 316L stainless steel is set to $0.2 \text{ N}/\mu\text{m}^3$). Through this process, it is possible to build a model with adequate numerical size to properly mesh the nanoscale coatings with fine enough elements. The element edge length of the coating is 25 nm while the element edge length of the substrate varied from 25 nm to $50 \mu\text{m}$. The total number of the elements for all models is approximately 400,000.

2.4. Boundary Condition and Load. The left edge of the model cannot translate horizontally, and the bottom edge of the model cannot translate vertically. The contacting surfaces are bonded, and no relative tangential translation would occur on the interface between them. In other words, all nodes of the neighbor surfaces on the interface that are at the same point at the initial stage will always stick together. Further experiments can be taken to analyze the actual behavior of the interface for more

accurate simulation. The whole section is linearly cooling down from 700°C to room temperature, 25°C , during the static structure simulation. Because the max temperature of the ITER enhanced flux first wall is about 800°C [22], the coatings analyzed in the previous study were annealed at 700°C for stress relief [21]. The mesh and boundary conditions of the finite element model are shown in Figure 3.

3. Result and Discussion

3.1. Al_2O_3 Coating and Y_2O_3 Coating. Stress fields of single-layer nanothick coatings with different thickness cooling from 700°C to 25°C are analyzed. As shown in Figure 4, the max principal stress in both Al_2O_3 and Y_2O_3 coating increases as the thickness increases when the thickness is below 500 nm. Moreover, as the thickness exceeds 500 nm, the max principal stress levels, showing little relevance to the thickness. The max shear stress in both coatings and the max Mises stress in Y_2O_3 show the same trend. And the max stress site of all these kinds of stress is located near the edge of the interface. The only exception is the max Mises stress in the Al_2O_3 coating where the max stress site is located in the center of the interface and the max value shows no relevance to the thickness.

The result reveals that there exists a critical thickness for a single-layer coating in which the exceeding values

FIGURE 2: The geometry of the model.

TABLE 1: Properties of the coating and substrate (25°C).

Materials	Density (kg/m ³)	Young's modulus (GPa)	Poisson's ratio	Coefficient of thermal expansion (10 ⁻⁶ K ⁻¹)	Thermal conductivity (W·m ⁻¹ ·K ⁻¹)	Specific heat (J·kg ⁻¹ ·K ⁻¹)
316L	8000 [19]	200 [19]	0.25 [27]	17.3 [19]	20 [19]	535 [19]
α -Al ₂ O ₃	3980 [19]	370 [19]	0.22 [19]	8.8 [19]	30 [19]	800 [19]
Y ₂ O ₃	5000 [28]	120 [28]	0.26 [28]	8 [28]	2.3 [24]	400 [28]

FIGURE 3: Mesh and boundary condition of the finite element ($t = 250$ nm).

contribute little to the residual stress. The edge of the interface can be considered a stress singular point because of the sudden change of the stiffness. When the stress singular region is small, the thermal residual stress at the edge, σ_p , can be expressed as follows:

$$\sigma_p = \frac{K}{r^\lambda}, \quad r < r_0, \quad (9)$$

where r_0 is the vicinity zone of the stress singularity and K is the stress intensity factor. Both K and λ are functions of Poisson's ratio and Young's modulus and the contact angles of the two layers. When the thickness of the coating

is small, with the thickness, t , increasing, the λ decreases, meaning that the stress singularity zone becomes wider and the σ_p at the edge increases [23]. When the thickness increases to a point, the bending effect which is negligible for a very thin film due to its very low stiffness plays a more dominant role. And the stress concentrated at the stress singularity will be relieved. According to Stoney's [24] equation, for the coating with enough thickness, the thermal stress in thin coating can be derived as follows:

$$\sigma_f = \frac{E_{ef} \int_{T_f}^{T_i} (\alpha_s - \alpha_f) dT}{1 + 4(E_{ef}/E_{es})(h/H)}, \quad (10)$$

(a) Effect of the thickness on the max principal stress

(b) Effect of the thickness on the max shear stress

FIGURE 4: Continued.

FIGURE 4: Effect of thickness on different kinds of stress in the system: (a) effect of the thickness on the max principal stress, (b) effect of the thickness on the max shear stress, and (c) effect of the thickness on the max Mises stress.

where $E_{ef} = E_f / (1 - \nu_f)$ and $E_{es} = E_s / (1 - \nu_s)$ are effective Young's modulus of the coating and the substrate, respectively. ν_f and ν_s are Poisson's ratio of the coating and substrate, respectively. h and H are coating thickness and substrate thickness, respectively. T_i and T_t are the initial temperature and terminal temperature. α_f and α_s are the thermal expansion coefficients of the coating and substrate, respectively. When the thickness of the coating is big enough, with the thickness increasing, the thermal stress decreases, because when the coating-substrate system is bent, bending-induced stress relaxation occurs. The thicker the coating, the more the stress reduction [25].

However, under the given condition in this simulation, the thickness-induced stress relief effect is still negligible even the thickness of the coating is 1000 nm. Meanwhile, 500 nm is the critical thickness for both Y₂O₃ coating and Al₂O₃ coating where the stress singularity effect on the increase in stress with the increasing thickness becomes negligible and the stress is almost not relevant to the thickness. It indicates that preparing coatings with more than 500 nm thickness will get consistent thermal shock endurance. It can make the quality control and the life span evaluation easier when the coatings are mass manufactured. But when the thickness of coating is below 500 nm, thinner coatings will have less residual stress and thus better thermal shock endurance.

On the other hand, all 3 kinds of stress in the Y₂O₃ coating are smaller compared with the Al₂O₃ coating at the same thickness because Young's modulus of Y₂O₃ is much smaller than that of the Al₂O₃, as their coefficients

of thermal expansion are almost even. Y₂O₃ coating is more deformable thus resulting in less residual stress in the system during thermal shock. Deformable material is favored when fabricating a deuterium permeation barrier exposed to thermal shock.

It should be noted that except the max Mises stress in the Al₂O₃ coating, the max stress site of all 3 kinds of stress of all models with varied thickness is located near the edge of the interface, as shown in Figure 5. The max Mises stress in the Al₂O₃ coating is located at the center of the interface. However, the local stress near the edge also concentrates near the interface. It reveals that the edge near the interface is the most likely crack initiation site with the most severe stress concentration in the system. It also shows that the stress distribution is more a geometry-relevant issue than a material property-relevant one.

3.2. Composite Coating. Previous work shows that the Y₂O₃/Al₂O₃ composite coating has excellent deuterium permeation resistance because the interface between layers contains defects that would trap deuterium and the lattice mismatch of 2 layers would cause the transmission mechanism of the deuterium to change [21]. In order to test the thermal shock endurance of this deuterium permeation barrier, the Y₂O₃/Al₂O₃/316L SS system was simulated using finite element analysis. The thickness of the Al₂O₃ layer is 150 nm which is close to that of the specimen as shown in Figure 1. The thickness of the Y₂O₃ top coating varies from 50 nm to 850 nm. As shown in Figure 4, the max stress in the Y₂O₃/Al₂O₃ coating was determined by the Al₂O₃ coating under these specific conditions. The max value of all 3 kinds

(a)

(b)

FIGURE 5: Continued.

(c)

(d)

FIGURE 5: Continued.

(e)

(f)

FIGURE 5: Comparison of 3 kinds of stress distribution: (a) max principal stress (Al_2O_3 , $t = 300$ nm); (b) max shear stress (Al_2O_3 , $t = 300$ nm); (c) max Mises stress (Al_2O_3 , $t = 300$ nm); (d) max principal stress (Y_2O_3 , $t = 300$ nm); (e) max shear stress (Y_2O_3 , $t = 300$ nm); (f) max Mises stress (Y_2O_3 , $t = 300$ nm).

of stress is almost equal to that of the 150 nm thick Al_2O_3 coating.

As shown in Figure 6, the max stress in the $\text{Y}_2\text{O}_3/\text{Al}_2\text{O}_3$ coating occurs in the Al_2O_3 layer and about an order higher

than that in the Y_2O_3 coating. Though the tensile stress of Y_2O_3 is not included in the CRC handbook, all tensile stresses of oxide ceramics listed in the CRC handbook are of the same order [26]. Thus, under the assumption that the tensile

(a) Al_2O_3 , $t = 150\text{ nm}$

(b) Y_2O_3 , $t = 200\text{ nm}$

FIGURE 6: Continued.

(c) $\text{Y}_2\text{O}_3/\text{Al}_2\text{O}_3$ (150 nm), $t = 350$ nm

FIGURE 6: Comparison of the max principal stress in 3 systems of the same individual thickness: (a) max principal stress in Al_2O_3 coating ($t = 150$ nm); (b) max principal stress in Y_2O_3 coating ($t = 200$ nm); (c) max principal stress in $\text{Y}_2\text{O}_3/\text{Al}_2\text{O}_3$ (150 nm) coating ($t = 350$ nm).

stresses of Y_2O_3 and Al_2O_3 are of the same order, the crack will originate from the boundary of the Al_2O_3 layer. Moreover, according to the result, stress distribution in each layer of the composite coating is similar to that of the monolayer coating of the same individual thickness. And coating an Y_2O_3 top coat over the Al_2O_3 layer have a little effect on the stress in the Al_2O_3 layer. And the max stress in the Y_2O_3 top coat is about 20% lower than that of the monolayer coating. The adjacent material, Al_2O_3 , has a more similar CTE to the Y_2O_3 than the 316L SS and causes less stress concentration at the edge. However, the Al_2O_3 is too thin to make such a big effect. The mesh quality of the adjacent layer also contributes to the decline of the max stress as the mesh of Al_2O_3 is much finer and regular than that of the 316L SS.

Adding an Y_2O_3 top coat have a little effect on the thermal stress in the Al_2O_3 . And the thermal shock behavior of the Al_2O_3 layer in the bilayer $\text{Y}_2\text{O}_3/\text{Al}_2\text{O}_3$ coating should be similar to that of the single-layer Al_2O_3 coating at the same thickness.

4. Conclusion

- (1) Y_2O_3 coating will introduce less residual stress under thermal shock compared with Al_2O_3 coating due to its smaller Young's modulus

- (2) The edge of the interface between the coating and the substrate is the most severe stress concentration site and thus the most likely crack initiation site in Al_2O_3 , Y_2O_3 , and $\text{Y}_2\text{O}_3/\text{Al}_2\text{O}_3$ coatings. And the max stress site and stress distribution pattern are more a geometry-dependent issue than a material property-relevant one
- (3) The residual thermal stress increases as the thickness of the coating increases, and the rate of the increase declines as the thickness increases due to the stress singularity effect. When the thickness of the coating reaches 500 nm, the residual thermal stress becomes less dependent on thickness
- (4) Adding an Y_2O_3 layer over the Al_2O_3 coating would not significantly affect the residual thermal stress in the Al_2O_3 coating. The thermal shock behavior of the Al_2O_3 layer in the $\text{Y}_2\text{O}_3/\text{Al}_2\text{O}_3$ composite deuterium permeation barrier could be evaluated as that of the single-layer Al_2O_3 coating of the same thickness of Al_2O_3

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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