

Microstructure optimization and hydrogen barrier mechanism of MAO/ZrO₂-Y₂O₃ composite coatings on zirconium hydride

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Abstract: In this study, by combining micro-arc oxidation (MAO) and sol-gel technology, the composite MAO/ZrO₂-Y₂O₃ hydrogen barrier coatings were successfully prepared on the surface of ZrH_{1.8} zirconium hydride matrix. The effects of the content of Y₂O₃ concentration (0-16 mol%) and heat treatment temperature (300-600°C) on the microstructure, phase composition, and hydrogen-blocking properties of the coatings were systematically investigated. The results showed that the composite coatings exhibited optimal performance when the Y₂O₃ content was 12 mol%, the defect area fraction was as low as 2.9%, the tetragonal phase of zirconia (t-ZrO₂) content reached 18.32%, the hydrogen permeation reduction factor (PRF) was elevated to 19.3, and the hydrogen-loss temperature was increased to 682°C. The heat treatment at 600°C further optimized the surface integrity and crystallization of the coatings, and led to the increase of the hydrogen loss temperature to 686°C. The hydrogen barrier mechanism of the coatings was attributed to the synergistic effect of physical barrier, chemical trapping of oxygen vacancies and phase stabilization.

Key words: hydrogen permeation resistance, sol-gel coating, microarc oxidation, ZrO₂-Y₂O₃ composite coating

ZrH is an ideal neutron moderator for miniature nuclear batteries due to its high hydrogen density, large macroscopic scattering cross-section and high temperature stability^[1-3]. Zirconium hydride possesses a low coefficient of thermal expansion (CTE), exhibiting minimal dimensional changes at temperatures up to 600°C, which ensures structural integrity in high-temperature environments. At the same time, its high thermal conductivity allows for efficient heat dissipation, which is essential for compact reactor design^[4]. Despite these advantages, ZrH is subject to decomposition at high temperatures, and this hydrogen desorption reduces the H/Zr ratio, decreases the neutron moderating efficiency, and promotes material embrittlement through lattice contraction, generation of

internal stresses, and increased susceptibility to microcracking^[3,5-7]. These hinder the further application of ZrH in miniature nuclear batteries.

Improvement of matrix components and surface modification techniques are effective means to prevent hydrogen escape. For example, alloying ZrH_{1.8} alloyed with elements such as Nb or Y can improve hydrogen retention by stabilizing the δ-phase, and the Niobium-doped ZrH_{1.6} can increase the hydrogen desorption temperature by about 50°C. However, over-alloying may reduce hydrogen density and neutron transparency^[8,9]. The formation of native ZrO₂ layers by in-situ oxidation can physically prevent hydrogen diffusion. Yan et al^[10] demonstrated that ZrO₂ layers grown in CO₂ at

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500°C can reduce hydrogen penetration by 60%. However, primary oxides are prone to cracking due to thermal mismatch with the substrate. Micro-arc oxidized can generate a dense zirconia layer (20–50 μm) by plasma discharge in an electrolyte. Thus, the MAO layer has a high hydrogen barrier potential due to its strong adhesion (bonding strength greater than 80 MPa) and high temperature stability^[11–13]. However, microcracks and monoclinic ZrO₂ (m-ZrO₂) phase transition inherent in the cooling process limit the hydrogen-blocking efficiency of the MAO film layers^[14]. Sol-gel coatings with high density can effectively block hydrogen escape through adsorption, making them a promising hydrogen barrier method^[15–18]. For example, Y₂O₃-doped ZrO₂ sol stabilizes the tetragonal ZrO₂ (t-ZrO₂) phase and inhibits crack extension and oxygen vacancy aggregation^[14]. Wu et al. reported that Si-Al/ZrO₂ sol-gel coatings reduced hydrogen permeability by 75% compared to MAO layers alone^[19]. Therefore, combining MAO with sol-gel technology can synergistically improve the hydrogen barrier properties of the coatings^[20]. However, the method of combining MAO with sol-gel still faces some limitations, such as the thermal expansion mismatch between the MAO layer and the sol-gel layer, which can generate interfacial stresses during thermal cycling and lead to the failure of the film layer. In addition, the components in the sol-gel may destabilize the ZrO₂ lattice and promote the formation of the cubic phase (c-ZrO₂), which results in a higher oxygen vacancy mobility and thus accelerates hydrogen diffusion^[21]. There are few studies on the methods of combining MAO and sol-gel, and a systematic correlation between sol-gel composition and hydrogen barrier properties is lacking. In addition, the heat treatment parameters have a great influence on the coating properties^[22], but the optimal heat treatment parameters are yet to be further revealed.

Therefore, in this study, zirconium oxychloride was used as a precursor to successfully construct an MAO/ZrO₂-Y₂O₃ composite hydrogen-blocking coating on the surface of ZrH_{1.8} matrix, and the preparation process was optimized. By systematically regulating the Y₂O₃ doping concentration (from 0 to 16 mol%) and the subsequent heat treatment temperature (300–600°C), the synergistic effects of these two key parameters on the coating microscopic morphology evolution, physical phase compositional features and hydrogen permeation behaviors have been investigated in depth. The multi-scale hydrogen barrier mechanism of the composite coatings was elucidated by combining field emission scanning electron microscopy (SEM), X-ray diffraction (XRD) and quadrupole mass spectrometry characterization.

1 Experiment

1.1 Materials

In this study, the zirconium hydride (ZrH_{1.8}) substrate was used, with the hydrogen-to-zirconium atomic ratio confirmed to be approximately 1.8. Prior to coating deposition, the bulk

material was precision-machined into disk-shaped samples measuring 20 mm in diameter and 2 mm in thickness using computer numerical control (CNC).

1.2 MAO treatment

Prior to micro arc oxidation (MAO), the specimens were subjected to a thorough surface preparation. First, the specimens were ultrasonically cleaned in acetone and ethanol, respectively, to remove any surface contaminants. Then, continuous mechanical grinding was performed using progressively finer silicon carbide sandpaper (200# to 1500# grit) to ensure a consistent surface integrity.

The MAO process was carried out using a T-MAO-B30 system with an electrolyte composed of sodium tripolyphosphate (Na₅P₃O₁₀, 18 g/L), disodium EDTA (Na₂EDTA, 1.5 g/L), and potassium hydroxide (KOH, 2 g/L). The electrical parameters applied during oxidation included a forward voltage of 430 V, a negative voltage of 120 V, a duty cycle of 50%, a frequency of 100 Hz, and a treatment duration of 15 min. Detailed electrolyte compositions and process conditions are summarized in Tables 1 and 2, respectively.

Table 1 The chemical composition of MAO electrolyte

Electrolyte	Na ₅ P ₃ O ₁₀	Na ₂ EDTA	KOH
Concentration (g/L)	18	1.5	2

Table 2 Micro-arc oxidation electrical parameters

Forward voltage/V	Negative voltage/V	Duty cycle /%	Oxidation time /min	Frequency /HZ
430	120	50	15	100

1.3 Preparation of the sol layer

The preparation processes of the composite coating are as shown in Fig. 1. Zirconium oxychloride was used as precursor to prepare the sol: zirconium oxychloride and yttrium nitrate were added into a mixed solution of anhydrous ethanol and deionized water with the mass ratio of 1:1, in which the molar ratios of Zr⁴⁺ to Y³⁺ were 100:0, 96:4, 92:8, 88:12, 84:16, respectively, and the solution was stirred with a magnetic stirrer to make a full and homogeneous mixing, and then H₂O₂ was added in which the molar ratio of H₂O₂ to Zr⁴⁺ was 3:1, and the reaction was heated and stirred for 30 min to make the reaction complete. Then add H₂O₂, in which the molar ratio of H₂O₂ and Zr⁴⁺ is 3:1, heat and stir for 30min to make the reaction complete, then slowly add ammonia to the solution to adjust the pH value of the solution to 3, and finally add 5% polyvinyl alcohol (PVA) solution to the solution to gradually form a transparent composite sol with the Tyndall effect, and then aged for 24h for preparation.

In this experiment, the sol-gel coating was applied by dip-

ping and lifting method. The specimen, which had undergone micro-arc oxidation, was immersed in the sol and lifted uniformly at a speed of 1.5 mm/s. This allowed the film to dry at room temperature until all the surface moisture and organic solvents had evaporated. The wet film layer was then obtained by repeating this process three times. After obtaining the wet film layer by impregnation and pulling, low-temperature drying pretreatment was carried out to exclude moisture and organic solvents from the sol, thus avoiding cracking of the film layer and generation of air holes due to solvent evaporation. The drying temperature and time were 80°C for 15 minutes, 50°C for 30 minutes, 30°C for 30 minutes and 260°C for 20 minutes. The above steps were repeated five times.

The heat treatment process was key to film formation. In this study, TG-DSC equipment was used for thermogravimetric–differential thermal analysis to investigate the influence of the heat treatment process on film quality. The equipment parameters were as follows: the heating rate was set at 10 K/min and the temperature range was 25–1000°C. Based on TG-DSC analysis, three heat treatment processes were designed: (1) heating to 300°C at 5°C/min and holding for 2h, (2) heating to 450°C at 5°C/min and holding for 2h, and (3) stepwise heating to 350°C at 2°C/min (holding for 1h), then to 500°C (holding for 1h) and 600°C at 5°C/min (holding for 2h). All samples were furnace-cooled.

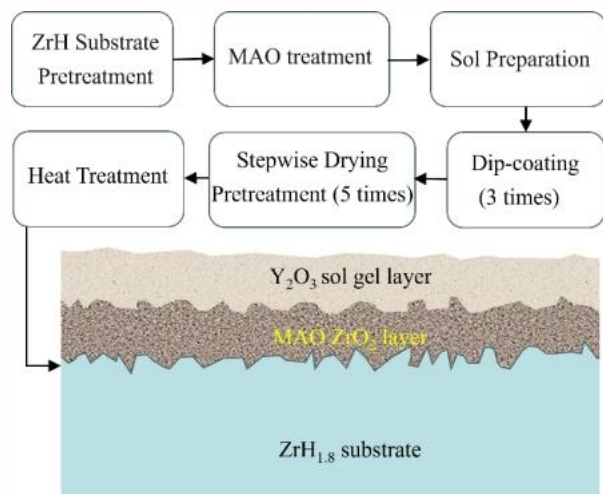


Fig. 1 Flow chart of MAO/ZrO₂-Y₂O₃ composite coating fabrication process

1.4 Characterization methods

The surface and cross-sectional morphologies of the composite film deposited on the zirconium hydride substrate were examined using a HITACHI S-6500 scanning electron microscope (SEM). The defect area fraction was determined by IPP software using a gray threshold of 100 based on average histograms, where white areas (defects) and black areas (dense regions) give the defect area fraction as white/total area ratio. Elemental distribution and composition of the specimens were

determined through micro-area point and surface scanning via energy-dispersive X-ray spectroscopy (EDS). Phase structure and composition analysis were performed using an ESCALAB-250Xi X-ray diffractometer operating at 40 kV and 35 mA, with a scanning rate of 3°/min across a 2θ range of 20°–80°. Film thickness measurements were obtained using a MiniTest 745 precision thickness gauge, while surface roughness was evaluated by averaging multiple area scans conducted with an OLS5000-3D laser scanning confocal microscope, which also captured three-dimensional topographical images of each film layer. Adhesion strength of the coatings was assessed through scratch testing employing a WS-2005 Automatic Scratch Tester.

Evaluation of the hydrogen barrier properties involved two distinct methodologies. The first approach involved high-temperature dehydrogenation weight loss method, with performance quantified through the hydrogen PRF. The second method employed HPR-20 EGA quadrupole mass spectrometry to track hydrogen evolution during thermal treatment, with results expressed in terms of hydrogen release rate and corresponding temperature.

For the high-temperature dehydrogenation experiments, specimens were placed in a quartz reactor and purged with argon for 30 minutes prior to heating to establish a protective atmosphere, which was maintained throughout the test. Following evacuation, samples were heated from ambient temperature to 750°C at 5°C/min in a KSL-1200-X furnace, held at this temperature for 10 hours, then furnace-cooled. The hydrogen permeation reduction factor (PRF) was calculated using the following equation: PRF = hydrogen loss of uncoated ZrH / hydrogen loss of coated ZrH, where the hydrogen loss of each sample was determined by weighing the sample before and after the dehydrogenation test. PRF is a dimensionless factor, and a higher PRF value indicates better hydrogen barrier performance.

In the quadrupole mass spectrometry measurements, the experimental setup involved coupling the tube furnace to the mass spectrometer via a tee junction, allowing continuous monitoring of evolved gases during heating. Under argon protection, samples underwent identical thermal treatment (5°C/min heating to 750°C, 10h hold, furnace cooling), with the resulting hydrogen evolution profile plotted as release rate versus temperature. Lower hydrogen release rates and delayed evolution temperatures corresponded to enhanced barrier properties.

2 Results

2.1 The influence of Y₂O₃ content

2.1.1 Surface morphology

Fig. 2 shows the surface morphology of MAO/ZrO₂-Y₂O₃ composite membrane layers with different Y₂O₃ contents. Fig. 2(f) presents the original surface morphology of the bare

MAO coating before sol-gel modification, which exhibits far more severe penetrating cracks and dense micropores, confirming the obvious surface defects of the pristine MAO substrate. It is observed that when the Y_2O_3 content is 0 mol%, there are obvious cracks and small holes on the surface of the membrane layer; when the Y_2O_3 content is increased to 4 mol%, the surface morphology is improved, but a small number of cracks and holes remain; when the Y_2O_3 content reaches 8 mol%, the surface of the membrane layer tends to be continuous and smooth, and the holes and microcracks are significantly reduced; when the Y_2O_3 content is increased to 12 mol%, the membrane layer surface tends to be continuous and flat, the surface of the membrane layer is smooth and dense, almost no cracks and holes, showing the best uniformity and continuity; however, when the Y_2O_3 content continues to increase to 16 mol%, small cracks show again on the surface. Above all, the surface quality of the composite coating was optimal when the Y_2O_3 content was 12 mol%.

The formation of cracks and holes in the coatings may originate from two factors: the defects existing on the surface of the initial MAO coatings and the residual stresses generated by drying and curing during the heat treatment process. Under the same preparation and heat treatment conditions, the difference in the number of cracks in coatings with different Y_2O_3 contents may be related to the bonding force and the matching of the coefficient of thermal expansion between the gel coatings and the MAO film layers. When the Y_2O_3 content was 12 mol%, the thermal expansion coefficients of the gel coatings and the MAO film layer were most matched, which effectively relieved the interfacial stress and obtained the optimal surface morphology.

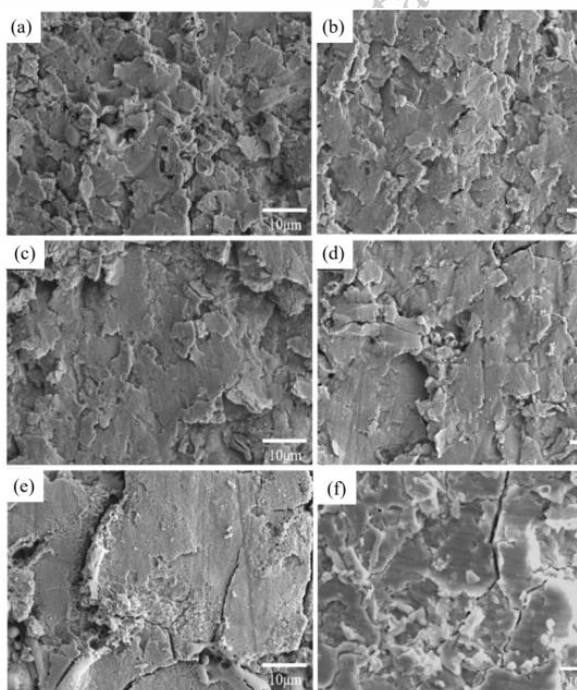


Fig. 2 Microstructure of composite coatings with different Y_2O_3

content: (a) 0mol%; (b) 4mol%; (c) 8mol%; (d) 12mol%; (e) 16mol%; (f) MAO

The laser confocal morphology of MAO/ ZrO_2 - Y_2O_3 composite film layers with different Y_2O_3 contents is shown in Fig. 3. It can be seen that the roughness of the composite coating gradually decrease with the Y_2O_3 content from 0 mol% to 12 mol%, the surface of the film layer becomes smoother and the surface quality gradually improves, while the surface roughness gradually decreases, as shown in Fig. 3 (f), whereas the surface of the composite layer exhibits a bumpy topography and the roughness is increased when the Y_2O_3 content is increased to 16 mol%. When the Y_2O_3 content is 12 mol%, the surface roughness is the lowest, which is $R_a=0.796\mu\text{m}$. The roughness test results show the same pattern with the surface morphology.

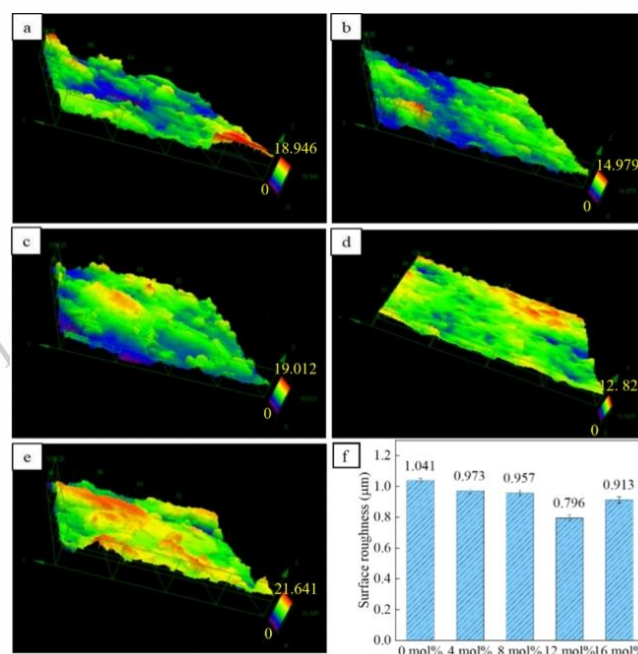


Fig. 3 Surface roughness of composite coatings with different Y_2O_3 content: (a) 0mol%; (b) 4mol%; (c) 8mol%; (d) 12mol%; (e) 16mol%; (f) surface roughness

2.1.2 Surface defect area fraction

The surface morphology of MAO/ ZrO_2 - Y_2O_3 composite coatings with different Y_2O_3 contents is shown in Fig. 4. It can be seen that with increasing Y_2O_3 content, the dense area (black area) gradually increases, while the defective area (white area), including both pores and cracks, gradually decreases. When the Y_2O_3 content exceeds 12 mol%, the pores and cracks in the composite coating slightly increase again, as shown in Fig. 4(e). The statistical results of the defect area fraction for the composite coatings with different Y_2O_3 contents are presented in Fig. 4(f), which reflects the extent of surface defects, including cracks and pores. With increasing Y_2O_3 content, the defect area fraction first decreases and then

increases. When the Y_2O_3 content reaches 12 mol%, the defect area fraction reaches a minimum of 2.9%, indicating that Y_2O_3 can stabilize the crystalline structure of ZrO_2 and reduce the formation of pores and cracks in the composite coating.

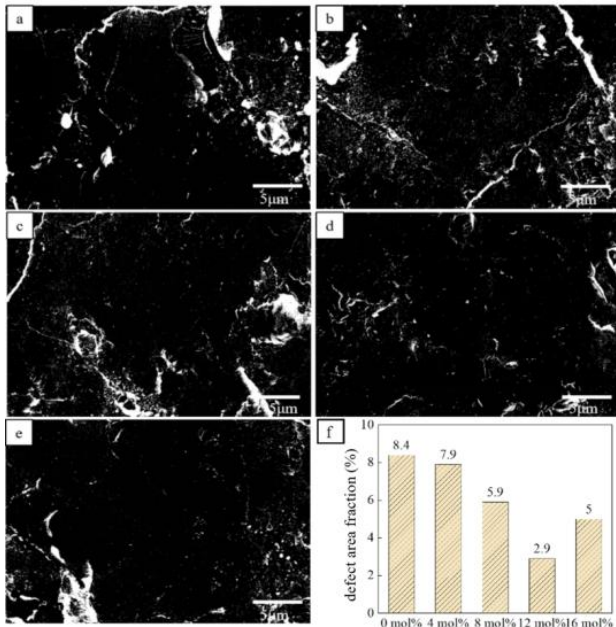


Fig. 4 Surface defect area fraction of composite coatings with different Y_2O_3 content: (a) 0 mol%; (b) 4 mol%; (c) 8 mol%; (d) 12 mol%; (e) 16 mol%; (f) statistical surface defect area fraction

2.1.3 Cross-sectional morphology

Fig. 5 shows the cross-sectional morphology of MAO/ ZrO_2 - Y_2O_3 composite coatings with different Y_2O_3 contents. The composite coating consists of a MAO layer and a sol-gel layer. The outer part of the MAO layer contains cracks and holes, while the inner part shows no obvious cracks. The thin sol-gel layer adheres well to the MAO layer without delamination, indicating good sealing. When the Y_2O_3 content is 0 mol%, the sol-gel layer is discontinuous, as shown in Fig. 5(a). With the increase of Y_2O_3 content, the densification of the sol-gel layer is improved. However, when the Y_2O_3 content increases to 16 mol%, a few of defects appear in the sol-gel layer and the densification decreases, as shown in Fig. 5(e). The MAO/ ZrO_2 - Y_2O_3 composite layer has the least defects and the best densification when the Y_2O_3 content is 12 mol%.

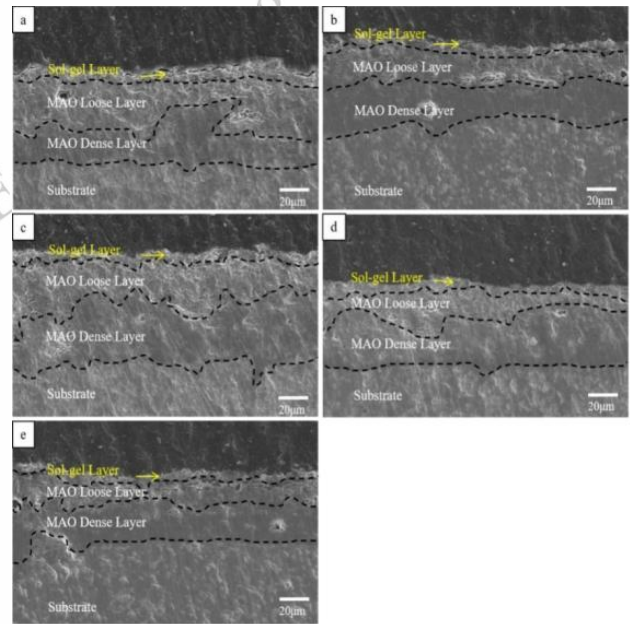


Fig. 5 Cross-sectional morphology of composite coatings with different Y_2O_3 content: (a) 0 mol%; (b) 4 mol%; (c) 8 mol%; (d) 12 mol%; (e) 16 mol%

2.1.4 Adhesion force

Fig. 6 shows the thickness and bonding force of MAO/ ZrO_2 - Y_2O_3 composite coatings. With increasing Y_2O_3 content, the coating thickness first increases and then decreases, ranging from 57 μm to 71.4 μm , reaching a maximum of 71.4 μm at 8 mol% Y_2O_3 . The bonding force exhibits a decreasing-then-increasing trend, with the lowest value observed at 8 mol% Y_2O_3 , followed by a gradual increase with further addition of Y_2O_3 . The bonding force is influenced by multiple factors, including coating composition, thickness, and interface roughness. The variation in coating thickness (57–71.4 μm) may also contribute to the observed changes in bonding force. Therefore, while Y_2O_3 content is considered a key factor, the bonding force results reflect the combined effects of composition, thickness, and interfacial mechanical interlocking^[23].

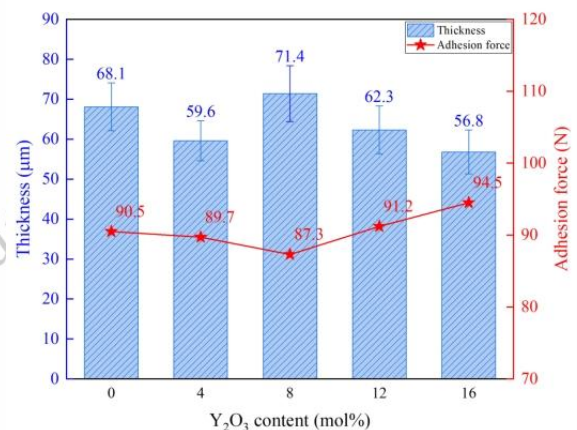


Fig. 6 The thickness and adhesion force of composite coatings with different Y_2O_3 content

2.1.5 Coating composition

Fig. 7 demonstrates the EDS results of MAO/ZrO₂-Y₂O₃ composite film layers. The results show that when the Y₂O₃ content increases to 4 mol%-16 mol%, the coexistence of the three elements, Zr, Y and O, is detected in the film layer. It is noteworthy that significant cracks exist on the surface of the samples without Y₂O₃ addition (Fig. 7a), but the number of cracks decrease significantly with the increase of Y₂O₃ content. It was observed that Zr and Y elements were distributed across the entire surface, including the cracked regions, with no clear evidence of preferential enrichment in the cracks (Fig. 7b-e). Further observation reveals that the uniformity of the distribution of Zr and Y elements gradually improved with the continuous increase of Y₂O₃ content.

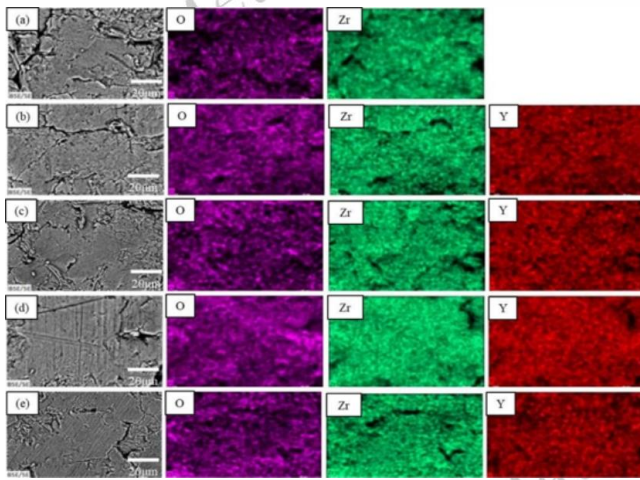


Fig. 7 Surface element distribution of composite coatings with different Y₂O₃ content: (a) 0mol%; (b) 4mol%; (c) 8mol%; (d) 12mol%; (e) 16mol%

Fig. 8 shows the XRD results of MAO/ZrO₂-Y₂O₃ composite coatings with different Y₂O₃ contents. All coatings are mainly composed of m-ZrO₂ (monoclinic) and t-ZrO₂ (tetragonal). With increasing Y₂O₃ content, the intensity of t-ZrO₂ diffraction peaks gradually increases, while that of m-ZrO₂ peaks decreases. This trend clearly indicates that Y₂O₃ promotes the formation and stabilization of the tetragonal phase and effectively inhibits the phase transformation from t-ZrO₂ to m-ZrO₂. When the Y₂O₃ content reaches 16 mol%, the diffraction pattern suggests the possible presence of cubic ZrO₂ (c-ZrO₂), indicating that the Y₂O₃ content has exceeded the critical value required for stabilizing the cubic phase. These changes in phase composition are expected to directly influence the mechanical properties and thermal stability of the composite coating, which are important factors for its potential application as a hydrogen barrier.

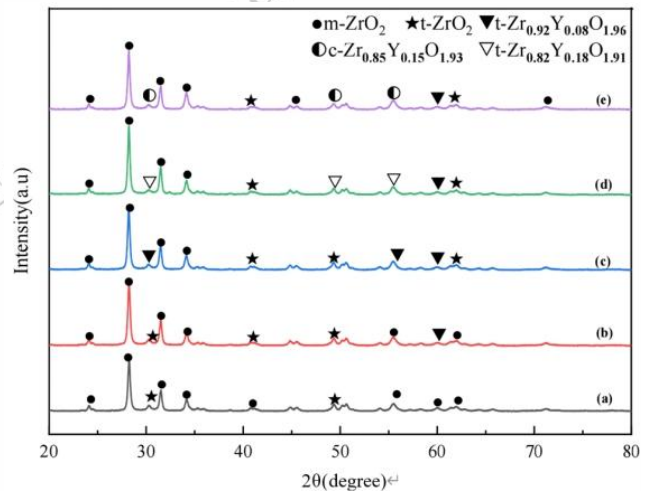


Fig. 8 The X-ray diffraction results of composite coatings with different Y₂O₃ content: (a) 0mol%; (b) 4mol%; (c) 8mol%; (d) 12mol%; (e) 16mol%

Table 3 and Fig. 9 shows the relative content of each phase in the MAO/ZrO₂-Y₂O₃ composite film layers with different Y₂O₃ contents. With the increase of Y₂O₃ content, the content of tetragonal phase increases and then decreases, and the content of monoclinic phase decreases and then increases, and the content of tetragonal phase is the highest and the content of monoclinic phase is the lowest when the Y₂O₃ content is 12 mol%. When the Y₂O₃ content reaches 16 mol%, the content of tetragonal phase decreases, which may be due to the transformation of part of tetragonal phase to cubic phase caused by the excess of Y³⁺ ions.

Table 3 The content of ZrO₂ phases in composite coatings with different Y₂O₃ content

Y ₂ O ₃ /mol %	0	4	8	12	16
m-ZrO ₂	85.83	85.09	82.94	81.68	82.16
t-ZrO ₂	14.17	14.91	17.06	18.32	17.84

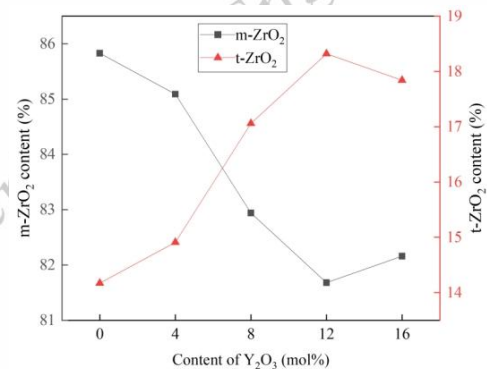
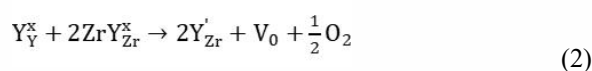
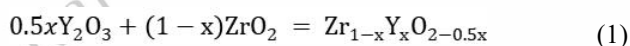


Fig. 9 The phase composition of composite coatings with different Y₂O₃ content

With the increase of Y content in the solid solution phase, the concentration of oxygen vacancies increases accordingly. Some related studies have shown that the generation of oxygen vacancies is the main factor inhibiting the crystallographic transformation of zirconia^[24-26]. The generation of oxygen vacancies is mainly due to the introduction of metal cations into zirconia, and the ionic radius of Zr^{4+} is 0.079 nm, while that of Y^{3+} is 0.089 nm, which is similar to that of Zr^{4+} , and the entry of Y^{3+} will not cause serious lattice distortion in zirconia. The entry of Y^{3+} into the crystal structure of zirconia replaces the position of Zr^{4+} to form a replacement solid solution, which produces oxygen defects in the crystal structure of zirconia, and the surrounding O^{2-} moves closer to Y^{3+} and farther away from Zr^{4+} , which reduces the lattice tension produced by the oxygen defects. The doping reaction formula and defect generation mechanism of ions are shown in Eq.1 and Eq.2.



2.1.6 Hydrogen barrier properties

Fig. 10 demonstrates the variation rule of hydrogen permeation factor (PRF) of MAO/ZrO₂-Y₂O₃ composite membrane layers with different Y₂O₃ contents. The data show that the PRF value shows a tendency of first increasing and then decreasing with the increase of Y₂O₃ content. When the Y₂O₃ content in the composite membrane layer is 0 mol%, the PRF value is the smallest 12.1, and when the Y₂O₃ content reached 12 mol%, the composite membrane layer showed the best hydrogen barrier performance, and the PRF value reached the maximum value of 19.3.

It has been reported^[27] that the PRF is closely related to the zirconia phase composition, and when more tetragonal phases are present in the zirconia film layer, the performance of the film layer is superior. When the composite coating does not contain yttrium oxide, the zirconia will spontaneously transform from tetragonal phase to monoclinic phase, and the volume effect generated during the phase transformation process leads to microcracks in the membrane layer, which provides a fast diffusion channel for hydrogen infiltration, resulting in a lower PRF value. With the addition of Y₂O₃, Y³⁺ ions replace Zr⁴⁺ to form a solid solution, while oxygen vacancy defects are generated, thus reducing the formation of cracks in the film layer. The tetragonal phase content in the film layer reaches the maximum value when the Y₂O₃ content is 12 mol%, which is attributed to the dense microstructure that significantly hinders the hydrogen penetration. However, when the Y₂O₃ content was further increased to 16 mol%, the excess Y³⁺ would lead to the transformation of part of the tetragonal phase to the cubic phase, and at the same time, it might cause

the lattice distortion, which in turn reduces the hydrogen barrier effect, and the PRF value slightly decreases to 18.7. This finding provides an important basis for the optimization of the optimal doping amount of Y₂O₃ in the hydrogen barrier coatings.

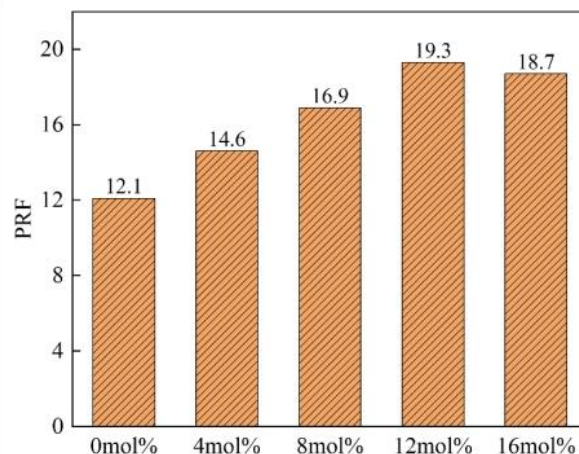


Fig. 10 The hydrogen PRF (high-temperature dehydrogenation weight loss method) of composite coatings with different Y₂O₃ contents

Fig. 11 shows the hydrogen release characteristic curves of MAO/ZrO₂-Y₂O₃ composite film layers with different Y₂O₃ contents. The hydrogen loss temperature of the specimen is different for different Y₂O₃ contents in the composite film layer. When the Y₂O₃ content is 0 mol%, the specimen loses hydrogen at 658 °C, and the composite film layer has no inhibitory effect on the hydrogen loss of zirconium hydride when the temperature continues to rise. With the increase of Y₂O₃ content, the hydrogen loss temperature showed a trend of increasing first and then decreasing, which are 4 mol% (665°C), 8 mol% (670°C), 12 mol% (682°C) and 16 mol% (675°C), and the specimen showed the best high temperature limit of hydrogen barrier when the Y₂O₃ content was 12 mol%. It can also be seen from Fig. 11 that the slope of the hydrogen release characteristic curve of the composite film layer containing Y₂O₃ is smaller than that of the composite film layer without Y₂O₃, which indicates the slower rate of hydrogen loss, and proves that the presence of Y₂O₃ slows down the rate of hydrogen loss of zirconium hydride. When the content of Y₂O₃ is 12 mol%, the slope of the composite film layer is the smallest, and the rate of hydrogen loss is the slowest.

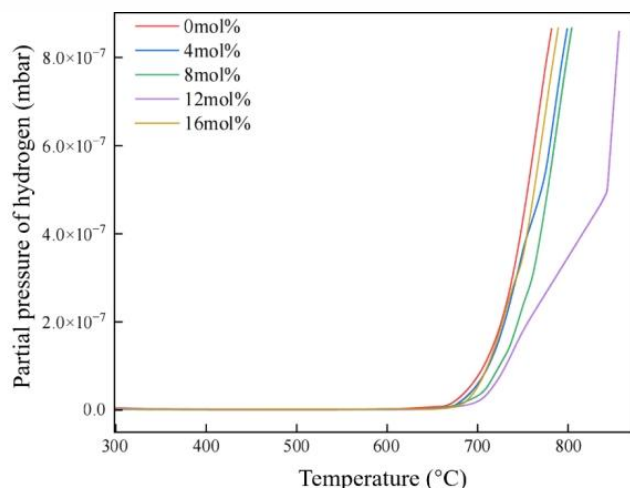


Fig. 11 The dehydrogenation curve (quadrupole mass spectrometry) of composite coatings with different Y₂O₃ content

2.2 The influence of heat treatment

The results of the above studies show that the MAO/ZrO₂-Y₂O₃ composite film layer with 12 mol% Y₂O₃ content exhibits the best hydrogen barrier properties. In order to further investigate the effect of heat treatment temperature on the performance of the film layers, the sol-gel with 12 mol% Y₂O₃ content was selected in this study and heat treated at 300°C, 450°C and 600°C to prepare the composite film layers, respectively.

It can be seen from Fig. 12 that the heat treatment temperature has a significant effect on the surface topography of the composite membrane layer. At 300°C, there are obvious cracks on the surface of the membrane layer with poor uniformity and continuity. When the temperature is increased to 450°C, the uniformity and continuity of the membrane layer are significantly improved, and the number of cracks is significantly reduced under the heat treatment condition of 600°C. This is because at the heat treatment temperature of 300 °C, the membrane layer crystallization is incomplete, only part of the amorphous state into the crystalline state, it is difficult to form a dense and continuous membrane structure. As the temperature increases, although the degree of membrane layer crystallization increases, the increase in the thickness of the membrane layer leads to the accumulation of internal stresses that may trigger cracking. When the temperature reaches 600°C, the organic matter and water in the gel are completely volatilized, the crystalline transition is complete, and a continuous and dense composite membrane layer is finally formed.

The surface defect area fraction test results further confirm this law, as the heat treatment temperature rises from 300°C to 600°C, the defect area fraction of the membrane layer decreases significantly from 8.6% to 1.9%. This trend is closely related to the phase composition of the membrane layer. At 300 °C, the membrane layer is dominated by monoclinic phase,

and the incomplete crystallization leads to more pores. At 450 °C, the appearance of tetragonal phase stabilizes the crystalline structure, and the pores are reduced; at 600 °C, the structure of the fully crystallized layer is the densest, and the defect area fraction is the lowest. These results fully demonstrate that an appropriate increase in heat treatment temperature can effectively improve the surface quality and microstructure of the composite membrane layer.

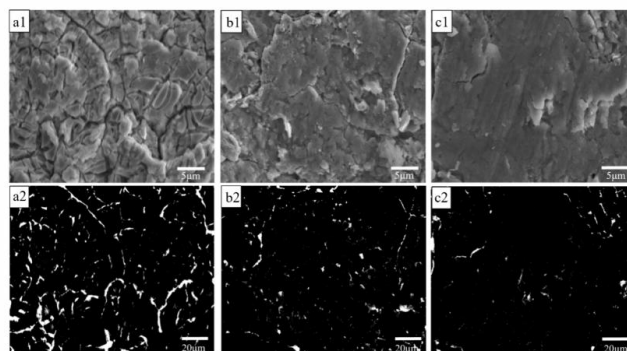


Fig. 12 Surface microstructure and surface defect area fraction of composite coatings after heat treatment at different temperature: (a1) and (a2) 300°C; (b1) and (b2) 450°C; (c1) and (c2) 600°C

Fig. 13 shows the XRD patterns of MAO/ZrO₂-Y₂O₃ composite coatings at different heat treatment temperatures. The coatings are mainly composed of m-ZrO₂ and t-ZrO₂. With increasing temperature, the diffraction peaks of t-ZrO₂ become stronger and sharper, while those of m-ZrO₂ gradually weaken. This indicates that a higher heat treatment temperature promotes the formation and stabilization of the tetragonal phase, which is beneficial for the hydrogen barrier performance of the composite coating. The absence of substrate peaks confirms that the coatings are thick enough to mask the underlying ZrH_{1.8}^[28].

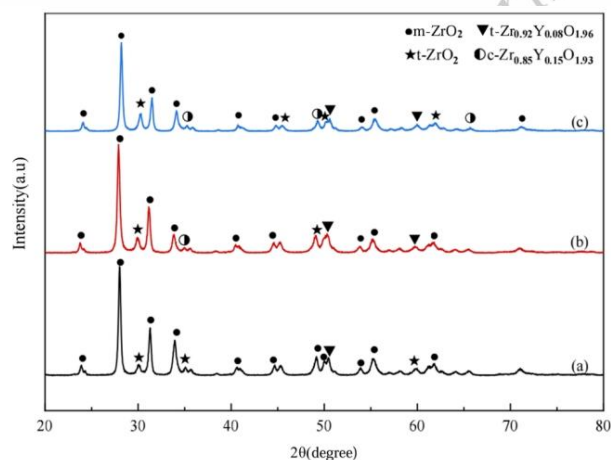


Fig. 13 XRD results of composite coatings after heat treatment at different temperature: (a) 300°C; (b) 450°C; (c) 600°C

Fig. 14 shows the hydrogen release characteristic curves of

MAO/ZrO₂-Y₂O₃ composite film layers at different heat treatment temperatures. It can be seen that the specimen heat-treated at 300 °C began to loss hydrogen at 652 °C. When the heat treatment temperature is increased to 450 °C, the hydrogen loss phenomenon occurs at 668 °C. As the temperature continues to increase to 600 °C, the hydrogen loss temperature increases to 686 °C. With the gradual increase of the heat treatment temperature, the hydrogen loss temperature of the specimen also gradually increased, and the hydrogen inhibition effect was gradually improved.

The hydrogen barrier effect of MAO/ZrO₂-Y₂O₃ composite film layer is related to the surface quality and phase structure. When heat treatment at 300 °C, the surface of the composite film layer is not uniform, there are cracks, the composite film layer consists of a large number of monoclinic phases, the surface defect area fraction of the composite film layer is 8.6%, the defects are more on the precipitation of hydrogen blocking effect is poor, so the beginning of the temperature of the hydrogen loss is low; at 450 °C heat treatment, the surface of the composite layer is relatively continuous, the tetragonal phase structure, the composite layer surface is 4.8%, the composite layer surface is relatively dense. At 600 °C heat treatment, the composite film layer formed is dense and continuous, the defect area fraction is 1.9%, and the composite film layer contains more yttrium-stabilized zirconia phase, the composite film layer has the best hydrogen blocking effect.

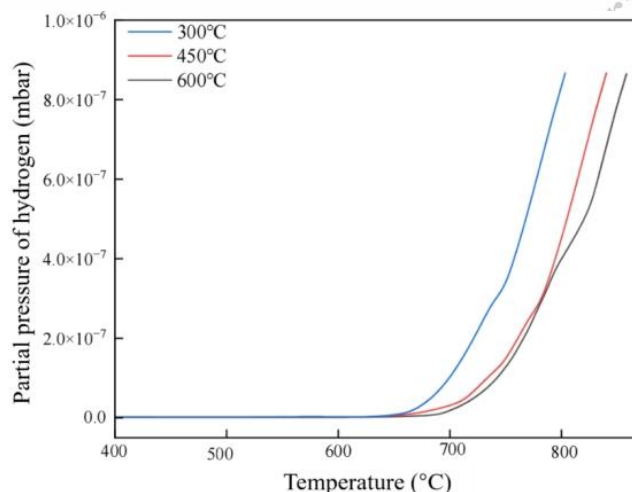


Fig. 14 The dehydrogenation curve (quadrupole mass spectrometry) of composite coatings after heat treatment at different temperature: (a) 300°C; (b) 450°C; (c) 600°C

Fig.1 shows the microstructures of the surface and cross section of SiC coated C/C composites. Fig.1a reveals that the surface of SiC coating has large cracks. The cracks have turned into the penetrating channel (shown in Fig.1b) through which oxygen can diffuse into C/C matrix and oxidize the matrix. Many holes exist in SiC coating simultaneously. At high temperature, these holes could shrink to destroy the coating.

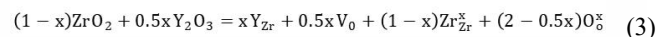
So, this structure of SiC coating is not beneficial to the anti-oxidation of C/C composites.

Fig.2 reveals the XRD patterns of SiC coating and TaSi₂ coating on the C/C composites. The new phases, TaSi₂ and Si, appear in the TaSi₂ coating comparing with the SiC coating. The rich Si phase in the coating plays the role of filling cracks due to its liquid state at the high temperature. According to the SEM image (Fig.3), no cracks exist on the surface of TaSi₂ coating.

3 Discussion

3.1 Mechanism of Y₂O₃ content on the properties of composite film layers

The results of this study showed that the introduction of Y₂O₃ significantly improved the microstructure and hydrogen barrier properties of the MAO/ZrO₂-Y₂O₃ composite membrane layer. When the Y₂O₃ content was 12 mol%, the membrane layer exhibited the lowest defect area fraction (2.9%), the highest tetragonal phase (t-ZrO₂) content (18.32%), and the optimal hydrogen permeation reduction factor (PRF = 19.3). This phenomenon can be attributed to the solid-solution effect of Y₂O₃. After the substitution of Zr⁴⁺ (0.089 nm) by Y³⁺ (0.089 nm), the lattice distortion is small due to the similarity of ionic radii, but the charge imbalance leads to the formation of oxygen vacancies (Eq. 3). These oxygen vacancies not only inhibit the phase transition from t-ZrO₂ to m-ZrO₂, but also reduce the hydrogen diffusion rate through the pinning effect^[29].



However, when the Y₂O₃ content exceeds 12 mol%, the film layer performance slightly decreases, which may originate from the oxygen vacancy clustering triggered by the excess Y³⁺, leading to the increase of the local lattice stresses and the increase of hydrogen diffusion channels. In addition, the EDS results in Fig. 7 show that the elemental distribution uniformity decreases at excessive Y₂O₃ content, which further validates the critical effect of doping concentration.

3.2 Effect of heat treatment temperature on film layer properties

The heat treatment temperature has a decisive influence on the densification and phase composition of the film layer. As shown in Fig. 15, the TG-DSC curve shows that the heat absorption peak at 120 °C is mainly due to the volatilization of free water and solvent in the gel powder. The heat absorption and exothermic peaks at 180 °C-220 °C are mainly due to the volatilization of the bound water in the gel powder and the dehydration of the precursor oxychloride. The heat absorption and exothermic peaks at about 260 °C-286 °C indicate that the

gel no longer contains the bound water and the bound water has been decomposed and accompanied by the decomposition of oxychloride. A sharp exothermic peak at 340 °C indicates that the gel skeleton decomposes and the oxides are generated from the oxychlorides and undergo the transition from amorphous to crystalline state. ZrO_2 gel undergoes the amorphous to crystalline transition at 450 °C, while $\text{ZrO}_2\text{-Y}_2\text{O}_3$ composite gel undergoes the crystalline transition at about 340 °C, and the Y_2O_3 content has an effect on the crystalline transition temperature. The content of Y_2O_3 has an effect on the crystalline transition temperature.

According to the TG-DSC curve, the thermal decomposition process of the composite gel was analyzed, and the optimal heat treatment process parameters were finally determined. At the initial stage, the gel contains a large amount of water, the heating rate should be slower, controlled at 2 °C/min. After the water in the composite gel is completely volatilized, the heating rate can be increased, controlled at 5 °C/min, the temperature can be increased to increase the sintering rate, controlled at 10 °C/min; and finally cooled down to the room temperature with the furnace.

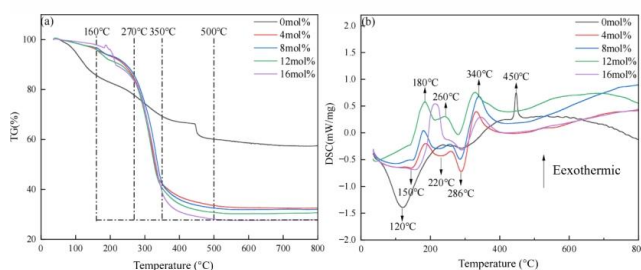


Fig. 15 TG-DSC curves of composite coatings: (a) TG curve; (b) DSC curve

The TG-DSC results show that the composite gel undergoes an amorphous-crystalline transition at 340°C, while the defect area fraction of the film layer decreases to 1.9% after heat treatment at 600°C with a significant increase in the content of t- ZrO_2 and c- ZrO_2 phases. This result correlates with the grain growth kinetics, where the increased rate of grain boundary migration at high temperatures contributes to the closure of pores, along with a more adequate diffusion of Y^{3+} , which further stabilizes the high-temperature phases.

It is noteworthy that the film layer treated under 300°C had a large number of cracks due to incomplete crystallization, leading to a hydrogen loss temperature of only 652°C. In contrast, the film layer treated under 600°C had a high hydrogen loss temperature of 686°C due to the dense t- $\text{Zr}_{0.82}\text{Y}_{0.18}\text{O}_{1.91}$ phase and low defect area fraction raised the hydrogen loss temperature to 686°C. This pattern is consistent with the study of Wu et al^[29], suggesting that heat treatment at the appropriate temperature is critical for optimizing the properties of the sol-gel layer.

3.3 Hydrogen blocking mechanism of composite film layers

The hydrogen-blocking performance of the composite membrane layer depends on the following synergistic mechanisms: firstly, the physical barrier effect, where the MAO dense layer (20-50μm) and the sol-gel sealing layer together reduce the hydrogen permeation pathway. The defect area fraction of the membrane layer with 12 mol% Y_2O_3 is only 2.9%, which is much lower than that of the composite membrane layer without Y_2O_3 . Secondly, it is the chemical trapping effect of the membrane layer, where the oxygen vacancies introduced by Y_2O_3 doping can adsorb hydrogen atoms to form O-H bonds and retard the bulk diffusion of hydrogen^[25,29]. Meanwhile, the XRD results showed a positive correlation between the t- ZrO_2 content and the PRF value, which verified the contribution of phase stability to the hydrogen barrier. Thirdly, the role of interface optimization of the composite film layer, by adjusting the Y_2O_3 content and optimizing the heat treatment regime, the difference in the thermal expansion coefficient between the MAO layer and the sol-gel layer can be reduced, which reduces the generation of interfacial cracks, and thus improves the bonding force, ensuring that no defects are generated in the film layer, and also guaranteeing the stability of the composite film layer in the harsh service environment.

Although the Y_2O_3 content and heat treatment temperature was optimized in this study, the following limitations still exist, for example, the long-term stability of the composite film layer has not been evaluated, and the effects of thermal cycling or stress cycling on the bonding and phase composition of the film layer need to be investigated. In addition, the hydrogen permeation mechanism is to be refined, and the interaction mechanism between oxygen vacancies and hydrogen can be further revealed by isotope tracing or first nature calculations. The threshold segmentation is sufficient for coating evaluation, future work could employ machine learning-based detection methods to monitor coating formation in real time and prevent defects during the MAO and sol-gel processes^[30-32], thereby enabling early-stage intervention in defect formation and significantly improving the densification and hydrogen barrier performance of the composite coatings.

4 Conclusions

The synergistic effects of the combined MAO and sol-gel $\text{ZrO}_2\text{-Y}_2\text{O}_3$ coatings on the surface integrity and hydrogen-blocking properties were investigated, the following conclusions were drawn:

1) The MAO/ $\text{ZrO}_2\text{-Y}_2\text{O}_3$ composite coating exhibited a dual-layer structure composed of a MAO layer and a sol-gel layer, which effectively reduced the number of surface cracks and pores. The incorporation of Y_2O_3 stabilized the tetragonal phase t- ZrO_2 , with its relative content reaching 18.32% at 12

mol% Y_2O_3 , significantly enhancing phase stability.

2) The optimal Y_2O_3 content was identified as 12 mol%, at which the composite coating achieved the lowest defect area fraction (2.9%), the highest hydrogen permeation reduction factor (PRF=19.3), and the most uniform elemental distribution. This composition minimized defects and optimized the hydrogen diffusion barrier.

3) Heat treatment temperature played a critical role in coating performance. A temperature of 600°C yielded the most compact and crystalline structure, reducing defect area fraction and increasing the hydrogen-loss onset temperature to 686°C, demonstrating superior thermal stability and hydrogen-blocking efficiency.

4) The hydrogen-blocking mechanism was attributed to the synergistic effects of physical barrier formation, chemical hydrogen trapping by oxygen vacancies, and phase stabilization. The performance of the composite coating was further validated by its high adhesion strength and resistance to thermal stress, making it a promising candidate for high-temperature nuclear applications.

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Author contributions

Investigation, conceptualization, initial drafting, Chen L W. Formal analysis, writing guidance and editing, Chen W D. Supervision, software, data treatment, Yan S F. Validation, writing—review & editing, Wang Y. Investigation, methodology, Cui X L. Software, data treatment, Zhou J Y. All authors have read and agreed to the published version of the manuscript.

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钎化氢表面 MAO/ZrO₂-Y₂O₃ 复合涂层的微观结构优化与阻氢机制

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摘要: 本研究通过将微弧氧化(MAO)与溶胶-凝胶技术相结合, 在ZrH_{1.8}氢化锆基体表面成功制备了MAO/ZrO₂-Y₂O₃复合阻氢涂层。系统研究了Y₂O₃浓度含量(0~16 mol%)和热处理温度(300~600°C)对涂层微观结构、物相组成及阻氢性能的影响。结果表明, 当Y₂O₃含量为12 mol%时, 复合涂层表现出最优性能: 缺陷面积分数低至2.9%, 氧化锆四方相(t-ZrO₂)含量达到18.32%, 氢渗透降低因子(PRF)提升至19.3, 氢损失温度提高至682°C。600°C热处理进一步优化了涂层的表面完整性和结晶程度, 使氢损失温度升高至686°C。涂层的阻氢机理归因于物理屏障、氧空位化学捕获和相稳定化的协同效应。

关键词: 阻氢渗透; 溶胶-凝胶涂层; 微弧氧化; ZrO₂-Y₂O₃复合涂层

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