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Chromia coating with nanofluid deposition and sputtering for accident tolerance, CHF enhancement

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ABSTRACT

In Fukushima accident, zirconium cladding was rapidly oxidized with high temperature steam, which ultimately led to hydrogen explosion. To overcome materialistic limitation, accident tolerant fuel (ATF) was suggested to improve safety response of nuclear power plants during accidents by modifying cladding surface with various coating materials. When chrome was coated on cladding surface, it showed fewer weight gain by high temperature oxidation compared to bare zirconium cladding. Chrome forms chrome oxide or chromia (Cr_2O_3) when oxidized, and this layer prevents further oxidation thus protecting inner material from oxidizing. However, previous studies indicated that implementation of chrome containing alloys have major drawbacks such as excessive coating thickness or degraded critical heat flux (CHF). Instead, direct coating of chromia was suggested in this study with the expectation of CHF enhancement compared to other chrome alloy coatings. Chromia nanoparticles were coated on nichrome wire surface with boiling deposition of chromia nanofluid. Another method was applying RF sputtering with chromia target. Chrome coating with DC sputtering were also tested for comparison. Verification of chromia coating was conducted by three steps: CHF measurement with wire pool boiling, high temperature oxidation in furnace to compare the oxidation resistance of specimens, and surface investigation. Surface characteristics investigation were conducted with measurement of contact angle by sessile drop method, capillary wicking height, and scanning electron microscope image. Experimental results show that chromia coating significantly increased CHF. Weight gain by oxidation indicate chromia nanofluid coating had improved oxidation resistance property.

1. Introduction

The chief interest of nuclear safety is to prevent the radioactive core's exposure to the public due to meltdown or outer wall damage. When loss of coolant accident (LOCA) or pump malfunction occurs, the coolant cannot control the core's temperature rise and this situation happened during Fukushima accident where all power systems including emergency supply went black (station black out). The fuel claddings were exposed to high temperature condition and caused zirconium, the main component of the cladding, to react with water and steam. The oxidation of zirconium produced hydrogen and oxidation heat, accelerating the process ($Zr + 2\ H_2O \rightarrow ZrO_2 + 2\ H_2$). Hydrogen stacked up inside the until hydrogen took place, damaging containment building and releasing radioactive materials.

In extreme conditions, material's property becomes major design limitations. In the case of Fukushima accident, the accelerated oxidation ability of zirconium at high temperature played a critical role. Consequently, after the unfortunate event, accident tolerant fuel (ATF) concepts have been widely studied. The major goal of ATF is to show improved safety response or mitigation during accidents to secure sufficient time to cope with while preserving or enhancing normal operating performance. One direction of ATF is modification of cladding surface with coating materials of metals or ceramics [1]. Many experiments were conducted comparing oxidation with various coating materials on zirconium surface and amongst them chrome based coating show promising results [2]. Weight gain by oxidation decreased 75% compared to bare specimen and surface investigation with scanning electron microscope (SEM) indicated formation of chrome oxide, or chromia (Cr_2O_3) layer on the outermost surface. This chromia layer acts as oxygen diffusion barrier, thus preventing further oxidation and for this reason, chrome is applied to many industrial alloys [3,4].

When cladding surface is modified, other factors should be considered such as compatibility to current cladding design, thermodynamic limitations such as critical heat flux (CHF), and reactivity related parameters such as neutron absorption cross

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Nomenclature

D	wire diameter (mm)
g	gravitational acceleration (m/s^2)
h	latent heat (J/kg)
I	current (A)
L	wire length (mm)
q″	heat flux (W/m^2)
V	voltage (V)

Greek letters

β	receding contact angle (°)
ρ	density (kg/m^3)
σ	surface tension (N/m)
ϕ	inclined heater surface (°)

Subscripts

c	critical
f	liquid
fg	liquid to vapor
g	vapor
R	reference resistance
S	specimen

section. In related study, when 310 stainless steel was coated on cylindrical zirconium cladding, chromia layer formed by oxidation was not thick enough to block oxidation at higher range of temperature [5]. This led to rupture of the cladding, suggesting that effective coating thickness for oxidation resistance could be overlarge. If coating becomes too thick, it is hard to implement on current NPP designs since increased cladding volume affects pressure drop and neutron economics. CHF is another important parameter in nuclear safety. CHF enhancement can mitigate rapid temperature increase and heater surface damage during severe accident condition. There are many studies aiming for CHF enhancement with surface modification in pool boiling experiment by using metallic or ceramic coating methods. However, investigation of chrome coating on stainless steel plate by electroplating shows degradation compared to bare surface and silicon carbide (SiC) coating [6]. This was due to formation of hydrophobic chrome surface, leading agglomeration of larger bubbles and interrupting liquid supply. Another study indicates no distinguishable change of CHF when chrome is coated on 316 stainless steel by physical vapor deposition (PVD) [7]. Although coating by this technique resulted in hydrophilic surface, there were no porous structures formed.

To preserve chrome's oxidation resistant property and overcome thickness and CHF issues, this study suggests alternative approach, direct implementation of chromia coating. As mentioned above, there are many studies that refer to combination of ceramic coating on metallic surface with variety of coating technique options. One method that has been research trend for over past decade is utilization of nanofluids [8,9]. Nanoparticles are mixed with base fluids such as water, mineral oil, and refrigerant. When surface is heated, vaporization of nanofluid occurs, depositing nanoparticle on the surface. The deposited nanoparticles form surface structures that affects boiling heat transfer phenomena. Like chromia, many types of ceramic materials were used as nanoparticles for CHF enhancement studies, due to their hydrophilic characteristics. Wu et al. [10] conducted pool boiling with titanium and silicon oxide surfaces and concluded that hydrophilic surfaces increases the CHF enhancement rate and the wettability is quantified by static contact angle. Aznam et al. [11] also reports CHF enhancement by nanoparticle deposition that increases wettability and porosity of the surface. Likewise, chromia nanoparticle was expected to enhance CHF when it was formed with nanofluid boiling deposition.

The major goal of this study is preliminary assessment of chromia coatings respect to CHF enhancement and oxidation resistance. CHF was measured with wire pool boiling in atmospheric pressure. Four types of coatings were tested: bare nichrome (80/20) wire, two types of chromia coated wire with different coating method, and chrome coated wire. Chromia nanofluid was prepared by chromia nanoparticle and distilled water. Its stability was confirmed by zeta potential measurement and pH were controlled to enhance stability. For chromia coating with boiling deposition of chromia nanofluid, coating parameters of heat flux and boiling time were varied to achieve optimized CHF enhancement. Surface characteristics were investigated to analyze the CHF results. Surface morphology was investigated using scanning electron microscope (SEM) image. Surface wettability of each test samples was measured with contact angle by sessile drop method. Capillary wicking height was also measured to compare the wettability of coated wires. After CHF experiment and surface investigation, weight gain by oxidation of bare nichrome wire and chromia coated wire were evaluated in high temperature (800 °C) condition.

2. Experimental setup

2.1. Formation and Stabilization of chromia nanofluid

Chromia nanoparticle with particle size less than 100 nm (Sigma-Aldrich) was used to form 0.1 and 0.01 vol% chromia nanofluid with distilled water as base fluid. However, the crude colloid suspensions were not proper for experimental use since visual observation led to conclusion that the nanoparticles showed sedimentation and agglomeration, indicating low stability. Therefore, stability was assessed with zeta potential with Zeta Sizer (Malvern). Zeta potential's magnitude indicates repulsive forces between adjacent particles in suspension, and when absolute value of zeta potential is over 30 mV, the suspension is considered to have moderate stability for experiments. From reference data, chromia suspension was expected to have highest stability at pH of 10 [12]. Thus, pH was modified to 10 by putting adequate amount of NaOH solution and additionally sonicated for 10 min. Before zeta potential measurement, to eradicate the IR drop, 0.01 mol/L of NaCl powder was added as supporting electrolyte. Table 1 lists zeta potential measurement results of three different samples, indicating modified suspension has sufficient stability, and 0.01 vol% chromia nanofluid were selected in the boiling experiment since it had highest stability.

2.2. Chromia coating with nanofluid boiling deposition

Nichrome wire with length (L) of 55 mm and diameter (D) of 0.5 mm was selected as bare surface. By utilizing pool boiling experimental apparatus in Fig. 1, boiling deposition with chromia nanofluid was conducted to form chromia coated wire. Rectangular glass basin was placed on hot plate and test specimens were connected to copper electrodes coated by Teflon. Chromia nanofluid was filled in the basin as base fluid with adequate height,