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Original Article

Development of Cr cold spray–coated fuel cladding with enhanced accident tolerance

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A R T I C L E I N F O

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A B S T R A C T

Accident-tolerant fuels (ATFs) are currently of high interest to researchers in the nuclear industry and in governmental and international organizations. One widely studied accident-tolerant fuel concept is multilayer cladding (also known as coated cladding). This concept is based on a traditional Zr-based alloy (Zircaloy-4, M5, E110, ZIRLO etc.) serving as a substrate. Different protective materials are applied to the substrate surface by various techniques, thus enhancing the accident tolerance of the fuel. This study focuses on the results of testing of Zircaloy-4 coated with pure chromium metal using the cold spray (CS) technique. In comparison with other deposition methods, e.g., Physical vapor deposition (PVD), laser coating, or Chemical vapor deposition techniques (CVD), the CS technique is more cost efficient due to lower energy consumption and high deposition rates, making it more suitable for industry-scale production. The Cr-coated samples were tested at different conditions (500°C steam, 1200°C steam, and Pressurized water reactor (PWR) pressurization test) and were precharacterized and postcharacterized by various techniques, such as scanning electron microscopy, Energy-dispersive X-ray spectroscopy (EDX), or nanoindentation; results are discussed. Results of the steady-state fuel performance simulations using the Bison code predicted the concept's feasibility. It is concluded that CS Cr coating has high potential benefits but requires further optimization and out-of-pile and in-pile testing.

1. Introduction

Current nuclear fuel systems for light water reactors (LWRs) are based on a combination of a slightly enriched UO_2 pellet and a cladding made of a Zr-based alloy. This system has been used for about half a century, and its safety and performance have been continuously improved and optimized. However, rare events have occurred in which this system does not behave as safely as desired. These events include the accident at the Three Mile Island, TMI-2 reactor in 1979 and the severe events at Fukushima Daiichi in 2011. After the Fukushima accident, the nuclear industry, utilities, and research and governmental organizations started R&D programs with the objective of development of a nuclear fuel system with enhanced accident tolerance or development of accident-tolerant fuels (ATF).

One of the widely studied ATF concepts is multicomponent cladding (also known as coated cladding). The cladding is considered a near-term technology that can be developed and employed in several years. The concept is based on modification of current Zr-based alloys, on which different protective layers are deposited. Many coating materials have been studied by different groups, including different deposition techniques. This article summarizes results of testing of Zircaloy-4 (Zry-4) cladding coated with chromium using cold spray (CS) technique. Similar cladding concepts are under development at CEA/AREVA [1], Korea Atomic Energy Research Institute [2], Westinghouse Electric Company [3], Czech Technical University [4], Ukraine [5], Institute for Energy Technology [6,7], and other institutes.

2. Materials and methods

2.1. Material and sample preparation

Chromium was chosen as a coating material mainly due to its extraordinary corrosion resistance, high melting point, good

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strength, high hardness, and good wear resistance. At the same time, chromium does not have a high neutron absorption cross section relative to natural elements such as nickel and is widely available at a reasonable cost. Owing to their similar mechanical and thermal properties, chromium is expected to be compatible with Zr-based alloys up to their eutectic temperatures, which are summarized in Table 1. Moreover, operating experience with chromium inside reactor cores already exists because control rods in some LWRs were plated with chromium to increase wear resistance; chromium is also an alloying element in core structural steels.

Standard commercially available annealed Zry-4 was used as a substrate. The composition of the substrate as provided by the supplier and confirmed by EDX was the following: 1.32Sn-0.21Fe-0.11Cr-0.13O-BalZr. The Zry-4 material was cleaned, descaled, and handled before different experiments following the ASTM standard [8].

2.1.1. *Cold spray process*

Zr-based substrate materials coated with chromium using various deposition methods have been widely studied [5,12,13]; however, the CS technique brings new questions and uncertainties to the ATF development process that have to be further investigated. The CS process involves the acceleration of micron-sized particles (~5–75 μm) in the form of commercially available powders that are carried at a high pressure (6.8 bar/100 psi–69 bar/1,000 psi) and sometimes in a heated gas stream (RT–1,200°C) in the solid state toward a suitable substrate, upon which the particles undergo tremendous plastic deformation [14].

Upon impact, the plastic deformation disrupts and breaks down surface oxide layers on both the powder and the substrate, leading to a metallurgical bond and mechanical interlocking [15]. Because the feedstock powder is deposited in the solid state, the microstructure is retained after deposition, with the exception of dynamic recrystallization due to high strain levels. The CS process is good for materials that can not only undergo high levels of strain with low energy input but are also sufficiently work hardened to obtain the desired strength. An impact-induced shock wave interacts with the expanding contact edge of an impacting particle [16], followed by adiabatic shear localization during the high-speed deformation of the particles on impact; this impact is characterized by localized temperature increases and strain concentration. Approximately 90% of the work of plastic deformation is converted to heat; the flow stress of most metals is sensitive to temperature, decreasing as temperature increases. During impact of the solid feedstock powder particles, the oxide layers on both the powder and the substrate surface are disrupted by "jetting" of the material and are partially removed, along with other impurities at the particle–substrate interface, exposing highly reactive "virgin" metal and inducing subsequent metallic bonding between particle and substrate material [17]. The feedstock powder is in intimate contact with the exposed substrate, forming an adherent metallurgical bond as a result of the severe plastic deformation during particle impact [18]. Therefore, it can be deduced based on empirical evidence that impact-induced shock wave and adiabatic shear, in combination with mechanical interlocking, serve as predominant bonding mechanisms in CS.

The bonding mechanism associated with CS is analogous to that of cold welding, which is another type of solid phase welding process in which bonding is also the result of plastic deformation of the metals to be bonded. Bonding occurs at the "critical impact velocity" when two surfaces under extreme pressure are forced together such that the surface oxide layers are disrupted and bonding takes place between the opposing virgin metal surfaces [19]. Evidence from single particle impact experiments shows that the critical velocity of specific materials and the phenomenon of jetting are closely linked to shock-compaction phenomena [20].

The VRC Gen III high-pressure CS system was used to produce all the specimens and was operated using helium as the accelerating gas. The feedstock powder was pure chrome (Fig. 1), produced by Exotech, USA, and had an average particle size of 44 μm, an oxygen content of 600 ppm, and a nano-hardness value of 5.1 GPa. A De Laval tungsten carbide nozzle with a circular exit was used for this study. It has a 0.068-inch (1.75 mm) throat, a 0.2-inch (5 mm) exit with a 6-inch (152 mm) expanding length, and a 10° converging section. The remaining process conditions are listed in Table 2. The as-coated surface without any additional treatment is shown in Figs. 2 and 3.

2.2. *500°C steam oxidation test*

Samples were first tested in low temperature steam to simulate in-reactor corrosion of the cladding material. The choice of the steam testing temperature results from a compromise between the

Table 1
Properties of substrate and coating materials at room temperature [9–11].

Material property	Chromium	Zircaloy-4
Ultimate tensile strength [MPa]	413	517
Yield strength [MPa]	362	348
Young's modulus [GPa]	140 (PVD coating)	99.3
Elongation[%]	0%	23%
Poisson ratio	0.21	0.37
Coefficient of thermal expansion [μm/m-°C]	6.20	6.00
Thermal conductivity [W/m-K]	69.1	21.5
Melting point/Eutectic [°C]	1860/1310	1850/1310
Specific heat capacity [J/g-°C]	0.461	0.285
Density [g/cm^3]	7.19	6.56
Crystal structure	bcc	hcp<~800°C < hcp + bcc<~1000°C < bcc
Thermal σ_a [barn]	3.1	0.2

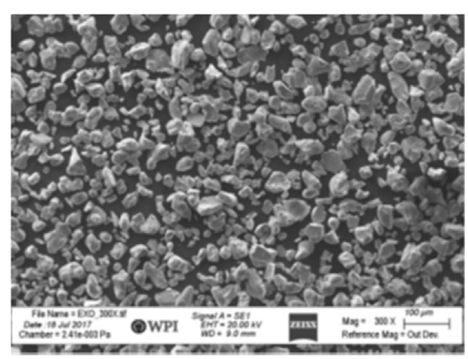

Fig. 1. SEM of the feedstock chrome powder produced by Exotech. SEM, scanning electron microscopy.

Table 2
CS process conditions used to deposit Cr on Zircaloy-4.

Nozzle type	WC-0.068
Carrier gas	Helium
Helium pressure [bar]	31.5
Preheater set point [°C]	700
Feeder set point	4
Standoff [inches]	1
Prechamber [inches]	3.5
Carrier gas flow [m^3/hr]	94
Helium flow [m^3/hr]	1319
Gun heater set point [°C]	675
Number of cycles	.25 (1 pass)
Gun speed [mm/s]	200