# Development of Nanostructured Ferritic Alloy (NFA) Fuel Cladding Tubes using Cold Spray Deposition Technology

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\*Keywords : Nanostructured Ferritic Alloy, Fast Reactor Fuel Cladding, Cold spray deposition technology

#### 1. Introduction

For sodium fast reactors (SFR) and lead fast reactor (LFR) nanostructured ferritic alloys (NFAs) are considered as the lead candidate materials for fuel cladding, providing superior creep strength and radiation resistance compared to conventional damage ferritic/martensitic (F/M) steels [1]. These superior properties are contributed to a high number density  $(10^{23})$  $-10^{24}/m^3$ ) of uniformly distributed nanometer-sized oxide clusters (2 - 10 nm) in a fine-grained (~ 0.5  $\mu$ m) ferritic steel matrix. Fig. 1 shows the typical microstructure and oxide nanoclusters in 14YWT (Fe-14Cr-3W-0.4Ti-0.2Y-0.250 wt.%, one of NFAs developed by Oak Ridge National Laboratory, ORNL). The oxide nanoclusters impede dislocation climb and glide, stablize the fine grain structure at elevated temperatures, and mitigate radiation-induced swelling and embrittlement on account of the large area of internal interfaces.



Fig. 1. TEM images showing microstructure of 14YWT and atomic probe tomography image of nanocluster in 14YWT [2].

However, the present fabrication methods for NFA cladding tubes for fast reactor designs are not amenable to large-scale manufacturing. The method involves consolidation of mechanically alloyed powders and

complicated thermo-mechanical steps such as hot extrusion (~ 400 °C), and multiple pilgering passes with intermittent annealing steps (850 °C – 1100 °C) for the shaping of cladding tubes [3]. To overcome this challenge, an innovative approach for manufacturing NFA cladding tubes using cold spray deposition technology (CST) was proposed and developed [3 – 5].

CST is a solid-state high velocity powder spray process, wherein the powder particles deform with very high strain rate  $(10^7 - 10^9 \text{ s}^{-1})$  upon impact with the substrate to form a thick dense deposit. CST is a commercial process and as such has a high technology readiness level (TRL). The proof-of-concept of manufacturing a free-standing ferritic steel tube by CST has been demonstrated.

The basic concept for the manufacture of NFA steel cladding tube using CST involves the deposition of NFA particles on a sacrificial aluminum-alloy tube mandrel and subsequent selective removal of the mandrel to leave behind a free standing NFA steel tube. Post-forming heat treatment is required for final densification and to facilitate the precipitation of nanoscale clusters. This manufacturing route is conducive to rapid factory manufacturing and has potential to overcome the limitations of current methods, in a cost-effective manner. Another phase of this study involves investigation of CST for deposition of functional coatings that would provide corrosion resistance for applications that crosscut a variety of current and advanced reactor concepts.

The present study evaluates the feasibility of CST manufacturing of NFA cladding tubes using two types of 14YWT powders (gas-atomized and ball-milled powders) in conjunction with microstructural characterization and mechanical tests. The effects of feedstock 14YWT powders on characteristics of final products such as grain structure, nanoparticle dispersion, and hardness are discussed.

## 2. Methods and Results

# 2.1. NFA Development using 14YWT Gas-atomized Powder

Initial feasibility of manufacturing NFA cladding tubes using the novel CST approach was demonstrated using gas-atomized 14YWT powders provided by Oak Ridge National Laboratory (ORNL). Extensive parametric study for CST including propelling gas type, gas preheat temperature, pressure, gun translational and tube rotational speeds, powder size distribution, and substrate material was performed. The powder deposition efficiency and density were significantly improved by using helium propellent gas and small size particles. Following deposition and removal of the underlying sacrificial mandrel, a free-standing NFA tubes were produced. It was then subjected to high temperature heat treatments at reducing environment. Fig. 2 shows a short length of the NFA cladding tube with a wall thickness of 1.2 mm and 200 mm length and flat samples.



Fig. 2. (Left) Photographs of an NFA tube and flat samples produced by CST and (Right) cross-sectional SEM image of the tube sample, exhibiting high-density microstructure.

The CST manufactured NFA samples heat-treated at 900 °C, 1000 °C, and 1100 °C were characterized to determine grain structure and orientation, and machined to small tensile samples (SSJ3 type). The as-deposited samples showed very fine grain structure (~200 nm) but brittle nature. However, annealing at 1100 °C increased the total elongation exceeding 30%. The grain size for the samples was about 5  $\mu$ m (Fig. 3). The yield strength and UTS of the samples was 320 MPa and 550 MPa, respectively, approaching the values for stainless steels.



Fig. 3. (a) Photographs of SSJ3 type tensile specimens and tensile testing facility, (b) EBSD data showing grain size distribution for NFA specimen annealed at 1100 °C, and (c) photographs of tensile samples after testing.

The NFA samples produced using gas-atomized 14YWT powder exhibited reasonable strength and good ductility while the grain size was much larger than that of 14YWT manufactured using the conventional method. This is mainly due to non-uniform dispersion of oxide nanoclusters in the NFA matrix. Therefore, the atomized powder should be subjected to severe plastic deformation by high-energy ball-milling to induce complete and

uniform solutionizing of all alloying elements prior to CST deposition.

#### 2.2. NFA Development using 14YWT Ball-milled Powder

The 14YWT ball milled powders were produced at Los Alamos National Laboratory (LANL). However, the ball-milled powders were not conducive to CST due to their high hardness and large size (i.e., a few hundred microns size). To make the balled-milled powders acceptable for CST a novel manufacturing route was developed. First, the ball-milled powders were cryomilled at liquid nitrogen to achieve smaller particle sizes by brittle fracture. To reduce the hardness, the powders were annealed in a mixture of hydrogen and argon environment at temperatures of 1000 °C and 1100 °C for 30 min. After then, the engineered powders were sprayed on stainless steel flats and Al-alloy flats. The deposition process produced dense thick deposits of NFA with 450 - 500 µm thickness. The produced flat samples and the cross-sectional SEM image of the deposit is shown in Fig. 4.



*Fig. 4. (a) Photograph and (b) cross-sectional SEM image of free-standing NFA flats produced using ball-milled 14YWT powders.* 

The annealed deposits up to 1100 °C exhibited a high-density microstructure with a high number density of nanoprecipitates. The hardness values of the annealed deposits were higher than bulk 14YWT produced by the conventional method. The result is an indication that the CST manufactured NFA using the ball-milled 14YWT has the desired mechanical properties. The major contributions to the high hardness values were originated from both grain boundary strengthening and dispersed nanoprecipitates.

#### 2.3. Multi-purpose NFA Designs

For applications in cross-cut multiple nuclear reactor concepts, depositions of coatings for corrosion and oxidation resistance using CST were investigated. Examples were CST process of FeCrAl (Fe20Cr5Al) and Cr coatings on the NFA materials. FeCrAl and Cr are highly resistant to high temperature oxidation and corrosion in primary water in LWRs. The developed materials are shown in Fig. 5. Air oxidation tests were performed with the coated samples at 1100 °C for 1 hour. The FeCrAl coated NFA developed only ~ 150 nm layer of Al<sub>2</sub>O<sub>3</sub> while uncoated sample revealed ~ 5  $\mu$ m film of Cr<sub>2</sub>O<sub>3</sub> on the surface. The experiment confirmed that functionally graded structural components can be

developed depending on reactor service condition using CST.



*Fig. 5. (Left) FeCrAl coated NFA flat and (Right) Cr coated NFA flat samples produced using CST.* 

#### 3. Conclusions

This study explored novel approaches to manufacture NFA material using cold spray deposition technology as an alternative to the conventional manufacturing route requiring arduous thermo-mechanical steps. After optimization of CST parameters, free-standing NFA cladding tubes with diameter and wall thickness prototypical to the actual fast reactor fuel cladding were successfully manufactured using as-atomized 14YWT powder. To achieve the desired microstructure and mechanical properties, ball-milled 14YWT powder was utilized for CST manufacturing. Powder engineering steps including cryomilling and annealing were used to make the powder conducive to CST. As a result, the produced NFA materials exhibited very fine grain size with uniformly dispersed nanoprecipitates as well as high hardness values. Finally, dual-material system consisting of FeCrAl (and also Cr) coated NFA material was developed for enhancing corrosion resistance of NFA for a variety of applications to cross-cutting reactor designs. The project was performed collaboratively by University of Wisconsin, Madison, Oak Ridge National Laboratory, Los Alamos National Laboratory, and Pohang University of Science and Technology in South Korea.

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