DEVELOPMENT OF ADVANCED COATINGS FOR NERVA-TYPE FUEL ELEMENTS

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Introduction: Nuclear thermal rocket (NTR) technology has been extensively investigated as viable alternatives to chemical rockets for deep space exploration mission in the United States since the 1950s under various programs starting with the Rover and NERVA programs, and currently under NASA's Nuclear Cryogenic Propulsion Stage (NCPS) program [1,2,3,4,5]. Before their termination in 1973, the Rover and NERVA programs built and ground tested over 20 nuclear reactor engines, and the overall technology development had attained NASA's TRL 6. Three types of graphite (Gr)-based fuel elements were developed and investigated in these programs [3]. Although uncoated graphite fuel elements were used in the Kiwi-A trials [3], subsequent engine tests used either NbC or ZrC-coated Gr fuel rods to minimize the reaction between the hot hydrogen and carbon. The use of coated fuels was demonstrated to increase engine life from 10 minutes in the early stages of the Rover program to 5 h towards the end of the program with a corresponding improvement in the performance of the engines. Despite impressive improvements in the quality of the coatings during the Rover and NERVA programs, "midrange corrosion" resulting in a significant loss in the mass of the fuel rods was a major issue affecting engine life and engine neutronics (Fig. 1) [2,3]. Cracks were observed in the coatings in the midrange area of the fuel rods due to a mismatch in the coefficients of thermal expansion (CTE) of the coatings and the matrix thereby enabling hydrogen ingress to the Gr in the fuel element matrix.

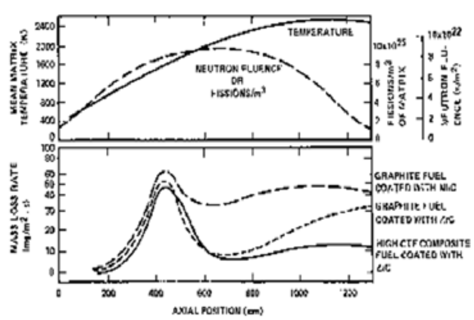

Fig. 1. Observed mass loss and reactor conditions along the axial lengths of Peewee and NF-1 fuel elements demonstrating the occurrence of midrange corrosion [3].

Proposed coating architecture: The present research proposes a new multilayered coating concept designed to solve the midrange corrosion problem. The concept envisions designing either a functionally graded or multilayered coatings to minimize the thermal expansion mismatch between the ZrC and the Gr/(U,Zr)C fuel matrix while acting as compliant layers as well as diffusion barriers separating the carbon and hydrogen. Noting that the chamber temperatures of the Rover and NERVA NTR engines varied between 2270 and 2695 K [3], and the requirements for using materials with a low thermal neutron absorption cross-sectional area, σ_a, it is useful to plot σ_a [6] against the absolute melting point, T_m, in order to identify suitable metallic elements (Fig. 2). An examination of Fig. 2 reveals that only Mo and Nb have $T_m \geq 2700$ K and $\sigma_a \leq 2.5$ barns. Since the CTEs of Mo and Nb are closer

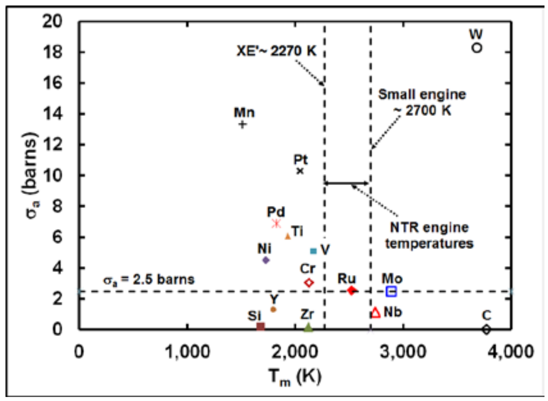

Fig. 2. Plot of thermal neutron absorption cross-section [6] against the absolute melting point.

to (U,Zr)C and ZrC, respectively, the thermal strains are expected to be reduced over the Rover legacy ZrC coatings. Preliminary elastic stress analyses of a coatings architecture such as that shown in Fig. 3 suggests that the tensile stresses on the ZrC coating would be considerably reduced over the legacy coatings (Fig. 4)

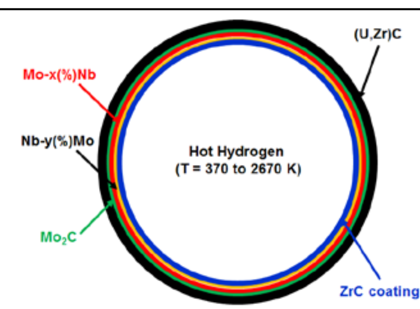

Fig. 3. Proposed coating architecture.

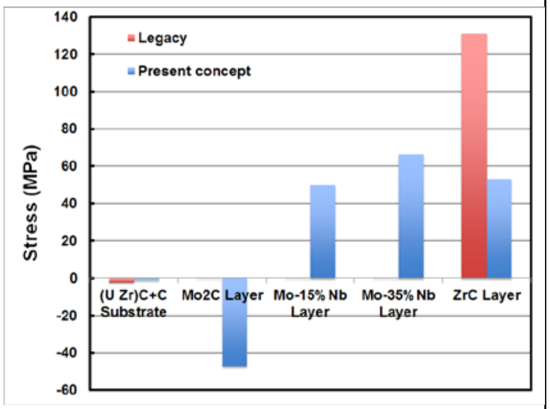

Fig. 4. Comparison of the elastic stresses developed in the legacy and proposed coating architectures.

[7]. It should be noted that the above analyses does not account for the expected plasticity of the Mo-Nb layers, which should further reduce the stresses. The fact that the Nb-Mo phase diagram is isomorphous with complete solid solubility [8] suggests that a functionally graded coating can be developed from an initially-deposited multilayer coatings by diffusion heat treatment. This complete solid solubility of these two metals allows for fine tuning the coating compositions for optimum performance. Additionally, Mo is less permeable to hydrogen diffusion [9], and the layers of Mo and Mo_2C (Fig. 3) are expected to act as barriers to carbon and hydrogen diffusion. Despite several potential advantages of the present concept, preliminary calculations show that increasing the Mo content would adversely affect the neutronics of engine designs using a thermal neutron spectrum [10]. Thus, it is essential to minimize the thickness and content of the Mo layer. Since significant diffusion will occur at the very high operating temperatures of NTR engines, it is also essential that the coating thicknesses be sufficient to minimize carbon and hydrogen diffusion during the mission cycle. Clearly, there will be a need to balance different requirements at some point in the design cycle if the present coating architecture is demonstrated to solve midrange corrosion.

The present investigation investigates the feasibility of using multilayered coatings to solve the midrange corrosion problem. A hot-pressed multi-layered $ZrC/Nb/Mo/Mo_2C/Gr$ specimen was thermally cycled between room temperature and 1873 K for 7 cycles. The layers did not debond after these thermal cycles despite the fact that each layer was more than 500 μm. Having demonstrated that the coating layers are well-bonded, the next step in the development process was to demonstrate that these layers can be deposited by chemical vapor deposition (CVD). Several CVD trial runs have been conducted at Technology Assessment & Transfer, Inc., Baltimore, MD.and the microstructures of the coatings are discussed. These trials have established that it is possible to deposit multilayered coatings by CVD although the quality of the coatings and the uniformity of their thicknesses require improvement. Experimental and theoretical studies are also underway to understand the diffusion of the various elements within the layers.

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