PROGRESS IN THE U.S. PROGRAM TO DEVELOP LOW-ACTIVATION STRUCTURAL MATERIALS FOR FUSION

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Abstract

It has long been recognized that attainment of the safety and environmental potential of fusion energy requires the successful development of low-activation materials for the first wall, blanket and other high heat flux structural components. Only a limited number of materials potentially possess the physical, mechanical and low-activation characteristics required for this application. The current U.S. structural materials research effort is focused on three candidate materials: advanced ferritic steels, vanadium alloys, and silicon carbide composites. Recent progress has been made in understanding the response of these materials to neutron irradiation.

1. INTRODUCTION

For fusion to be a viable energy source of the future it must offer safety and environmental advantages not found in current energy production options. One key advantage is the fact that the fusion reaction does not produce long-lived radionuclides or reactive chemical wastes. The structural components of a fusion power plant can become radioactive, however as the result of nuclear transmutations caused by exposure to neutron irradiation. The level of induced radioactivity in these structures is the predominant factor controlling the environmental impact of a fusion power plant. As a consequence, structural materials with low-activation characteristics are necessary to fully exploit the environmental potential of fusion. The U.S. Advanced Materials Program (AMP) is engaged in a science-based effort of theory, modeling and experiment to develop structural materials that will permit fusion to attain its safety, environmental and economic competitiveness goals. The objectives of the AMP are to 1) understand the behavior of candidate material systems in the fusion environment, 2) identify limiting properties and approaches to improve performance, 3) develop materials with acceptable properties through control of composition and microstructure, and 4) provide the materials technology required for production, fabrication and power system design. Currently, three materials systems are being investigated: advanced ferritic steels, vanadium alloys, and silicon carbide composites. Significant progress has been made toward understanding the performance of these materials under fusion relevant conditions.

2. ADVANCED FERRITIC STEELS

Advanced ferritic steels possess several characteristics which make them attractive candidates as fusion structural materials. They have high thermal stress resistance due to low thermal expansion coefficients and higher thermal conductivities than austenitic steels. They exhibit good corrosion resistance in water and liquid metals, and oxidation resistance at ~550°C. Processing and fabrication techniques are at a mature state for these materials because of extensive industrial experience, and because of substantial research conducted to develop these steels for use as cladding and duct materials in liquid metal reactors. Finally, these steels have demonstrated excellent swelling resistance under fission reactor irradiation conditions.

The advanced ferritic steels being considered for fusion are a modified composition of conventional Fe - 9-12% Cr steels [1]. Low-activation characteristics are obtained by removing elements such as Mo, Nb, Ni and specific impurities. In the low-activation steel Mo is replaced by W or V and Nb by Ta. The major technological challenges facing successful application of these steels include; 1) potential interaction of a ferromagnetic material with the high magnetic field of a fusion plant, 2) the effect of irradiation on fracture behavior, 3) the effect of transmutation generated He on mechanical properties and 4) relatively low high-temperature creep strength. The U.S. effort on advanced ferritic steels is part of an International Energy Agency cooperative program in which the U.S. role is to characterize the fracture behavior of these alloys.

Development of fusion structural materials requires detailed information on the fracture behavior of candidate structural materials to avoid unexpected catastrophic failure by unstable propagation of cracks. A material's effective toughness is a measure of its resistance to fracture. Fracture toughness always depends on the microstructure and properties of a particular material, but it will also depend on other variables such as loading rate, specimen geometry and size. Fracture toughness values, determined by standard test methods, yield an intrinsic material property only under very restricted conditions that are seldom met in practice. Standard fracture toughness measurement methods are not adequate for fusion power systems because the underlying principles of elastic-plastic fracture mechanics are violated for small specimens or in thin-walled structures with shallow cracks. Any fracture property database developed for nuclear systems must be created with relatively small specimens compatible with available irradiation volumes. Further, fusion first-wall structures will, by necessity, be thin-wall due to thermal stress limits. Thus, new approaches to characterize the effective fracture toughness as a function of temperature, $K_e(T)$, are required.

A goal of the U.S. program is to develop physically-based, micro-mechanical models of fracture to define the transition from quasi-cleavage to microvoid coalescence fracture modes. This is a general approach that seeks to determine the effective toughness by direct measurement of the sequence of events involved in the fracture process. Confocal microscopy (CM) and fracture reconstruction (FR) techniques are being developed [2] to examine crack blunting, tearing and damage development ahead of the crack tip. Confocal microscopy is used to obtain digital topographic maps of conjugate fracture surfaces from a test specimen. Fracture reconstruction is begun by computationally aligning the digital topographs so that the flat pre-crack surfaces are matched and overlapped until the position of the pre-crack front is accurately depicted. The surfaces are then computationally separated by an amount corresponding to the crack tip opening displacement, δ. A critical crack tip opening is reached when crack propagation is observed. This critical crack tip opening is related to the effective fracture toughness. To validate the CM/FR method a series of experiments was carried out using a variety of alloys, specimen configurations, loading rates and test temperatures to determine the effective toughness by both conventional methods and by CM/FR. A comparison of K_δ values determined from CM/FR with values of K_{Ic} determined from mechanical testing is plotted in Figure 1. Note this methodology provides a robust means to evaluate the effects of strain rate, irradiation, specimen size and constraint on K_e ($=K_\delta$).

The information obtained from CM/FR when coupled with finite element calculations of local crack tip stress fields can be used to determine the critical conditions for cleavage fracture. Critical stress-critical area (σ^*/A^*) models have been developed to predict the cleavage transition in small specimens for a variety of alloys, test types and specimen configurations [2]. These models can be used to predict $K_e(T)$ as a function of size and to directly adjust $K_e(T)$ data to a common test geometry. The power of this approach is illustrated in Figures 2 and 3 which compare uncorrected $K_e(T)$ data

(Figure 2) for an advanced ferritic steel against constraint corrected data (Figure 3). The curve in Figure 3 is the "Master Curve" for reactor pressure vessel (RPV) steels referenced at an effective toughness of 100 MPa√m and –75°C. The master curve is an alternative, perhaps simpler, approach that is also being developed to account for the effects of irradiation, strain rate, and specimen geometry and size. This method assumes that a master $K_e(T)$ curve exists that can be indexed to an absolute temperature scale by a reference temperature at a reference toughness. Changes from the reference state are reflected by adjusting the position of the master curve by an appropriate set of temperature shifts [2]. This approach has gained wide acceptance in fracture assessments of light-water RPVs.

Recent headway has been made in evaluating the effect of helium on fracture behavior. To achieve helium production rates prototypic of fusion in a mixed-spectrum reactor requires simulation techniques. One approach is to add isotopes of Ni to the steel. About 2% Ni is sufficient to give a He to dpa ratio representative of a fusion neutron spectrum. One-third scale Charpy impact specimens irradiated at 400°C to 40 dpa in the High Flux Isotope Reactor at ORNL suggest that He levels from 30 to 110 appm may act synergistically with hardening caused by displacement damage and precipitation to cause an increase in the ductile-to-brittle transition (DBTT) [3]. The change produced by He may be larger than that attributable to displacement damage or precipitation alone. Investigators in Japan and Europe, using different techniques such as B doping and cyclotron irradiations have observed similar effects. It should be noted that these results are not unequivocal, however, due to uncertainties associated with the Ni doping technique. Segregation of Ni to grain boundaries could cause atypical He accumulation that could lead to reduced fracture strength. This is an area requiring further research to determine the efficacy of the Ni doping method for He simulation.

3. VANADIUM ALLOYS

Vanadium alloys containing 4-5% Cr and Ti exhibit physical, thermal, and mechanical properties that are favorable for fusion applications [4]. These alloys are particularly attractive in combination with lithium-cooled blanket designs. Favorable characteristics include low long-term radioactivity, high heat load capacity and resistance to void swelling. The unirradiated tensile properties and limited thermal creep data suggest an upper temperature limit between 650 and 750°C. The industrial experience and manufacturing capacity of vanadium alloys is very limited in comparison with advanced ferritic steels, however scale-up from laboratory heats to commercial production of 500 and 1200 kg ingots has been successfully accomplished.

Early research in the U.S. on vanadium alloys indicates that Ti is effective for suppressing irradiation-induced swelling. Titanium also increases the tensile strength and improves the fabrication characteristics, but degrades creep strength. The addition of Cr to the alloy tends to increase the tensile and creep strength, and improves corrosion resistance in some cases. Irradiation-induced transmutations of V, Cr and Ti largely decay to the same three elements and all exhibit low long-term radioactivity. Thus, an alloy based on these three elements will exhibit minimal composition changes with high dose.

The presence of non-metallic impurities such as O, N, C and H can significantly effect the mechanical properties. These impurities can be introduced to the alloy during melting, fabrication processes, joining operations, service in partial vacuum or gaseous environments, and by mass transfer in liquid metal cooled systems. The effect of these impurities includes increased yield strength, loss of tensile ductility and decreased fracture toughness. The magnitude of the effect strongly depends on the distribution of the impurity element(s) in the microstructure. Evaluation of the performance of V-Cr-Ti alloys after exposure to environments containing O and H has been an active area of research within the U.S. program. The oxidation kinetics of V-(4-5)Cr-(4-5)Ti as a function of temperature in air and various oxygen partial pressures has been determined [5,6]. The data indicates that the oxidation process follows parabolic kinetics in all of the environments investigated. The temperature dependence of the parabolic rate constants is described by an Arrhenius relationship. The activation energy for oxidation in oxygen partial pressures ranging from 10^{-6} to 10^{-1} torr was fairly constant, but was significantly lower in air and substantially higher in pure oxygen. Tensile tests performed after pre-oxidation at 500°C showed that changes in mechanical properties were predominantly effected by the pre-exposure time and largely unaffected by oxygen partial pressure [6].

A major focus of the U.S. research and development effort for vanadium alloys is to explore the effects of irradiation on constitutive behavior and fracture properties at low-temperatures. A series of experiments has recently been completed at temperatures ranging from ~100 to ~500°C and doses between 0.1 and 18 dpa. Post irradiation examinations have not been completed, but preliminary results indicate significant changes in mechanical properties at temperatures below about 400°C [7]. Figures 4 and 5 [8-17] show the effect of irradiation temperature and dose on yield strength and uniform elongation for results obtained from the recently completed experiments together with data collected at higher irradiation temperatures and doses. Results for irradiation temperatures up to 600°C and doses to 49 dpa are included in Figures 4 and 5 to illustrate the transition in behavior above ~400°C. It is evident from the data that V-4Cr-4Ti exhibits rapid hardening and loss of strain hardening capacity following neutron irradiation at temperatures up to ~400°C. Large increases in yield strength are observed with concomitant decreases in uniform elongation. These changes in tensile properties are generally accompanied by large increases in the DBTT [7]. Microstructural analysis revealed that these property changes are related to a high density of small defect clusters which can be easily sheared by dislocations during deformation. Such a microstructure leads to flow localization or dislocation channeling which is responsible for the reduced uniform elongation [7]. These experiments have contributed substantial new information toward understanding radiation damage effects which govern the low-temperature operating limit for V-4Cr-4Ti. Further work is needed at temperatures between 300 to 500°C and in the low dose regime to accurately determine the minimum acceptable operating temperature for vanadium alloys in a fusion power system.

At high temperatures (≥ 650°C) the primary concern is the effect of neutron damage and helium transmutation on creep rupture properties. As with advanced ferritic steels the combined effects of neutron damage and helium generation can cause loss of tensile and creep ductility by growth and coalescence of helium bubbles at grain boundaries. Considerable experience has been obtained on V-4Cr-4Ti irradiated in the range of 420 to 600°C to neutron doses of 24 to 32 dpa, and with helium generation rates in the range of 0.4 to 4.2 appm/dpa. In this environment the material shows promising resistance to radiation-induced embrittlement and swelling. The DBTT did not increase above -100°C and density changes did not exceed 0.5% [18]. However, considerable work remains to be conducted at higher He and dpa levels, as well as at higher temperatures. To support future irradiation experiments at higher temperatures work has been initiated to explore the thermal creep properties of V-4Cr-4Ti from 600 to 800°C in vacuo and liquid Li. Tests performed in liquid Li will also investigate the influence of He on creep properties by tritium doping.

4. SILICON CARBIDE COMPOSITES

Silicon carbide (SiC) composites are attractive for structural applications in fusion energy systems because of their low-activation and afterheat characteristics, excellent high-temperature properties, corrosion resistance and reasonable dimensional stability. A helium-cooled SiC ceramic breeder blanket design is proposed in the ARIES concept [19]. With this concept upper use temperatures of 900 to 1000°C may be possible with the attendant benefit of high thermodynamic efficiency.

High-strength, continuous fibers with near stoichiometric SiC composition reinforce the composite and impart improved fracture toughness compared to monolithic SiC. The fibers are embedded in a SiC matrix produced by chemical vapor infiltration or infiltration and pyrolysis of a preceramic polymer. An interphase layer is deposited on the fibers prior to forming the matrix to permit debonding of the fiber-to-matrix interface as the composite is loaded. The matrix is generally of lower strength than the fibers and its primary function is to transfer load to the fibers while the debond layer prevents catastrophic failure.

Utilization of these new materials for fusion applications will require optimization of their radiation stability, thermal conductivity, gas permeability, chemical compatibility with fusion relevant environments, and joining methodology. Very little is known about the radiation performance of these materials, so the recent focus of the U.S. research program has been to explore the effects of irradiation on composite dimensional stability, strength and thermal conductivity. The primary issue is the development of a radiation tolerant SiC fiber and the integration of that fiber into an acceptable composite architecture.

It is well known that crystalline beta SiC exhibits good radiation stability. Linear swelling saturates at a relatively low dose and decreases with irradiation temperature from about 0.8% at 200°C to almost zero at 1000°C. In contrast, fibers irradiated in this temperature regime shrink with increasing radiation dose as shown by the data in Figure 6 [20]. The exception is Sylramic^{TM} which swells like crystalline SiC. Fiber shrinkage has been attributed to crystal growth and loss of oxygen [21,22]. Fibers fabricated from polymer techniques are not fully crystallized, but are composed of nano-scale crystallites embedded in a matrix of SiCO. Nicalon-CG and Tyranno^{TM} fibers both exhibited considerable radiation-induced shrinkage and contained high levels of oxygen relative to Hi-Nicalon and Sylramic fibers. Further, the crystallite size of the fibers displaying shrinkage was almost two orders of magnitude smaller than Sylramic (1.3 to 4 nm versus 85 nm).

Irradiation experiments on standard, commercially available SiC composites have demonstrated that a significant reduction in strength occurs at radiation doses as low as one dpa [23]. Irradiation experiments to higher doses have shown that the strength reduction tends to saturate at relatively low doses [24]. Strength reductions have been attributed to the poor irradiation performance of the Nicalon-CG fibers comprising the standard composite. As noted above, these fibers shrink and densify under irradiation. In addition, their strengths also decrease. As a result, the bend strength of composites made from these fibers is degraded by both fiber strength loss and de-coupling from the matrix. Recent high fluence (up to 80 dpa) results [25] confirm the trends observed at lower doses in that the strength of SiC composites decrease rapidly with increasing fluence initially, but attains a plateau of about 300 MPa at 800°C.

The thermal conductivity of SiC or SiC composites is significantly reduced by irradiation-induced point defect generation. For exposures to about 26 dpa the thermal conductivity of the standard composite was reduced by 60% from 7 to 3 W/m-K at 800°C. Monolithic SiC shows similar behavior under irradiation decreasing from about 36 W/m-K to around 26 W/m-K at 800°C. Recent advances in fiber and composite manufacturing techniques have demonstrated the capability to produce material with much improved thermal conductivity of 35 W/m-K at 1000°C [26].

Newly developed, advanced SiC fibers such as Hi-Nicalon, Nicalon-S and Sylramic as well as composites made from these fibers, are being produced and evaluated for fusion applications. These composites offer the potential for improved performance under neutron irradiation through increased radiation stability. The new fibers also display better thermal stability and creep strengths relative to Nicalon-CG [27]. New fiber/matrix interphase layers are also under development since the standard interphase material, carbon, lacks radiation and chemical stability. Composites with advanced interphases such as a defective or quasi-porous SiC layer, and a multi-layer interphase consisting of thin alternating layers of SiC/C/SiC are being developed. The multi-layer approach does not completely eliminate carbon from the system but reduces the quantity substantially relative to the conventional composite.

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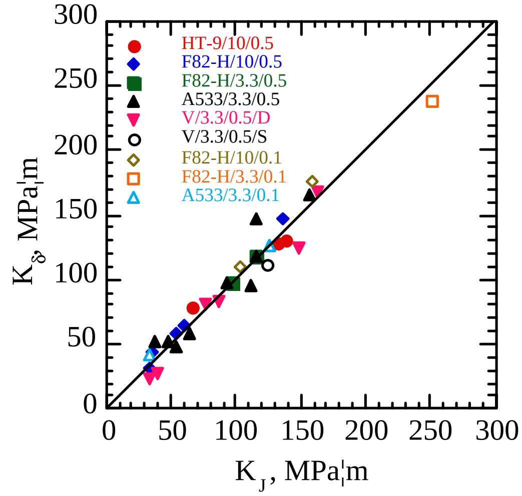

Figure 1 Comparison of K_δ values determined from CM/FR with K_{Jc} values determined in mechanical tests for a variety of materials and test conditions [2].

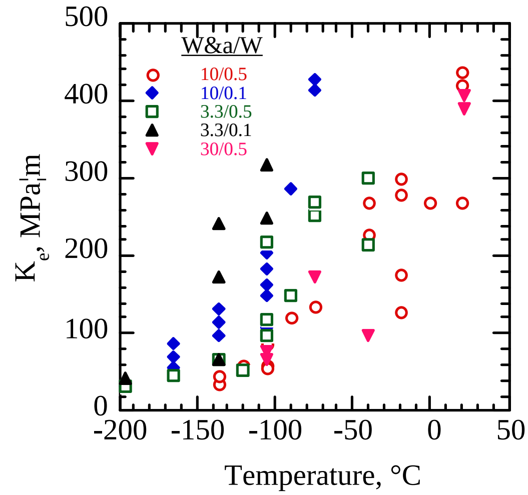

Figure 2 Static $K_e(T)$ data for an advanced ferritic steel from pre-cracked Charpy type specimens [2].

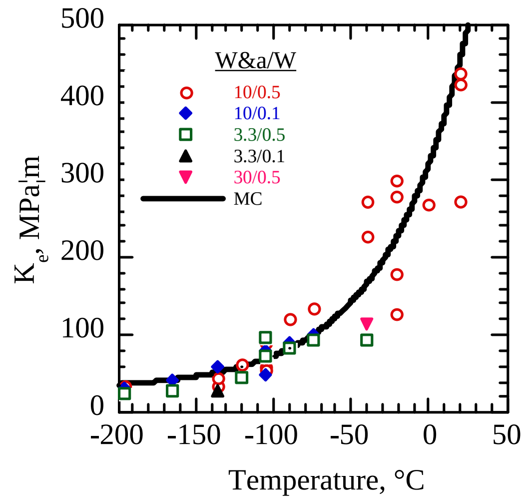

Figure 3 $K_e(T)$ data constraint corrected to a common reference specimen geometry [2].

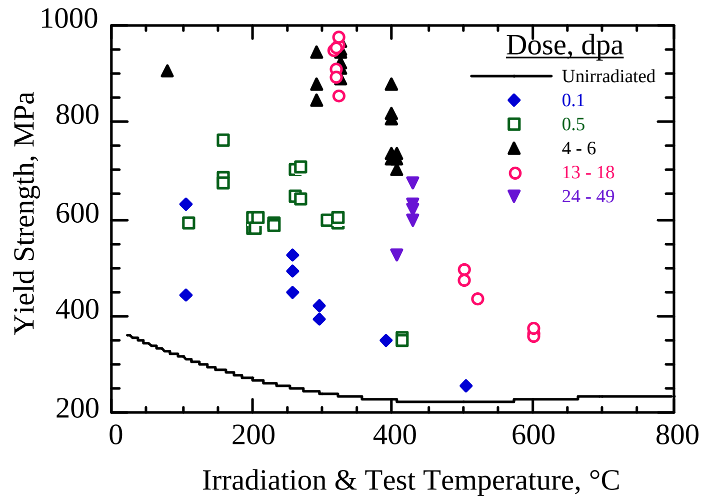

Figure 4 Yield strength of unirradiated and irradiated V-(4-5)Cr-(4-5)Ti alloys [8-17].

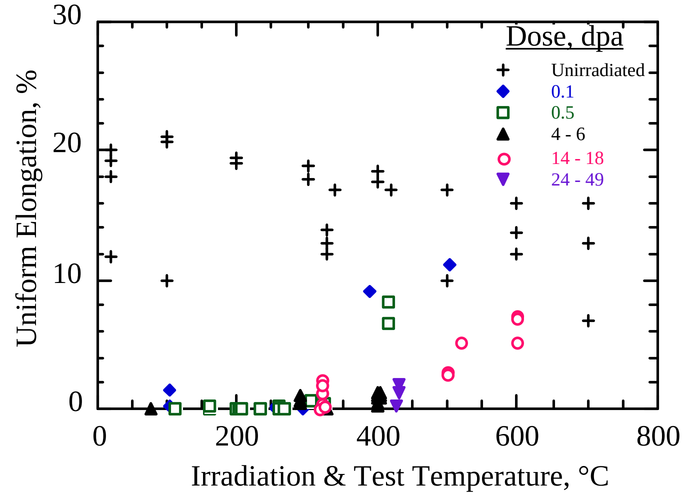

Figure 5 Uniform elongation of unirradiated and irradiated V-(4-5)Cr-(4-5)Ti alloys [8-17].

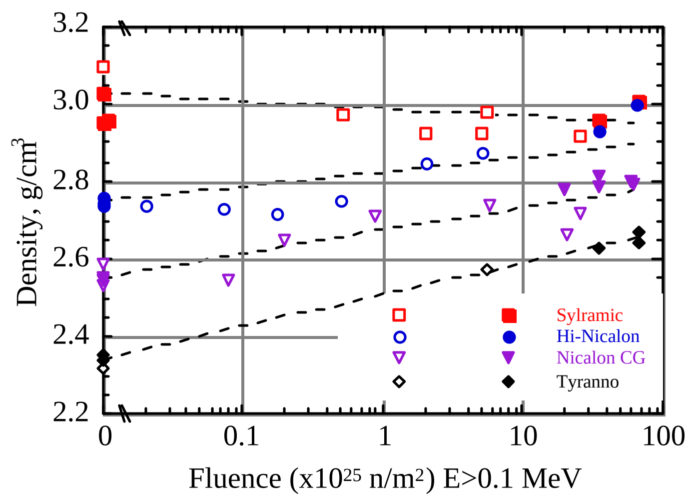

Figure 6 Effect of irradiation on density of SiC-based fibers [27].