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Materials degradation in fission reactors: Lessons learned of relevance to fusion reactor systems

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Abstract

The management of materials in power reactor systems has become a critically important activity in assuring the safe, reliable and economical operation of these facilities. Over the years, the commercial nuclear power reactor industry has faced numerous 'surprises' and unexpected occurrences in materials. Mitigation strategies have sometimes solved one problem at the expense of creating another. Other problems have been solved successfully and have motivated the development of techniques to foresee problems before they occur. This paper focuses on three aspects of fission reactor experience that may benefit future fusion systems. The first is identification of parameters and processes that have had a large impact on the behavior of materials in fission systems such as temperature, dose rate, surface condition, gradients, metallurgical variability and effects of the environment. The second is the development of materials performance and failure models to provide a basis for assuring component integrity. Last is the development of proactive materials management programs that identify and pre-empt degradation processes before they can become problems. These aspects of LWR experience along with the growing experience with materials in the more demanding advanced fission reactor systems form the basis for a set of 'lessons learned' to aid in the successful management of materials in fusion reactor systems.

1. Introduction

Thirty years ago, the requirements of fission and fusion systems were largely non-overlapping in terms of their challenges and their environments. Table 1 summarizes the principal issues facing a maturing commercial fission reactor industry and an evolving realization of requirements for materials in proposed fusion devices. Light water reactors faced fuel performance limitations due to Zircaloy creep, growth, corrosion and pellet–clad interaction, a newly arisen problem of intergranular stress corrosion cracking of stainless steel BWR pipes, the emergence of IGSCC in alloy 600 steam generator tubes, and embrittlement of copper – containing welds in reactor pressure vessel steels. Conversely, the fusion community had identified its own set of challenges; creep and fatigue of first wall materials, dimensional stability at high temperature and high doses, degradation of the first wall and plasma by sputtering, erosion, blistering, etc., induced activity in structural materials and the challenges in breeding tritium and in extracting it for future use. While core materials in fission reactors faced an environment

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Table 1
Fission/fusion materials challenges, circa 1975

Fission	Fusion
Zircaloy corrosion	First wall fatigue
Cladding creep and growth	Creep strength
Pellet-clad interaction (PCI)	Dimensional stability at high temperature and doses
BWR pipe cracking	Induced activity
Steam generator tube cracking	First wall sputtering, blistering, erosion
RPV embrittlement	Breeding materials
Very low capacity factors	Tritium retention and extraction

characterized by 320 °C temperatures, neutron doses up to ~10 dpa and low He production, fusion system analogues were required to withstand temperatures above 500 °C, doses over 100 dpa and high helium loading of plasma facing materials.

Today, uprating and life extension of commercial LWRs, demands by advanced fission reactor systems in the Generation IV initiative, [1] and the emergence of accelerator driven transmutation concepts [2] have pushed the environments of fission systems to higher temperatures, higher neutron fluxes, greater fluences and significantly more helium loading. In fact, as shown by Bloom et al. [3], the environmental conditions of advanced fission systems and magnetic confinement fusion systems are converging in most all aspects. In essence, both communities are facing very similar challenges in materials performance.

2. Key factors and processes governing materials performance

There are numerous important processes, environments and interactions that impact the performance of materials in a reactor system. The factors presented in Table 2 and discussed in the following are distinguished by either their sensitivity, their frequency of occurrence or their unanticipated consequences in the overall performance of the materials or material system.

Table 2
Key factors and processes governing materials performance

Temperature
Dose rate
Composition–microstructure
Coupling of factors/processes
Welds
Surface condition
Gradients
Incubation
Metallurgical variability
Environment (design)

2.1. Temperature

While temperature is an important factor in most materials degradation processes, it is the sensitivity of degradation to temperature that is sometimes surprising in fission reactors. An example is shown in Fig. 1, which is a dose-temperature plot of void formation in an austenitic alloy irradiated in the BN-350 reactor [4]. Because measurements were taken along the length of a component in the core, the data covers a wide temperature range but are confined to a narrow dose range for that component (identified by *strings* of data). Also, since the *strings* of data come from the same component, uncertainties caused by metallurgical variability are eliminated. The filled data represent cases where swelling occurred and the open data represent cases where swelling was not observed. There is a striking dependence on temperature in that *all* samples above 307 °C exhibited swelling whereas *none* of the samples below 302 °C swelled. This is a very narrow temperature threshold delineating the swelling from the non-swelling regimes, extending over a dose range of 3–57 dpa, and demonstrates the extreme sensitivity of swelling to temperature. Similar, but not as dramatic, examples exist for swelling in baffle bolts in pressurized water reactors (PWR), [5] and in stress corrosion cracking of steam generator tubes [6].

2.2. Dose rate

Equally striking are dose rate dependencies observed in fission reactor systems. For the *same* dose, a reduction in dose rate by a factor of ~10 can result in an increase in irradiation hardening in model pressure vessel steels, by as much as 30%, Fig. 2 [7]. Similarly, for 304 stainless steel irradiated in EBR-II, a reduction in flux by a factor of 20 has been observed to produce a *decrease* in the incubation dose for swelling from ~20 dpa to ~5 dpa [8].

2.3. Composition and microstructure

Composition and microstructure can have profound effects on material behavior, and in some

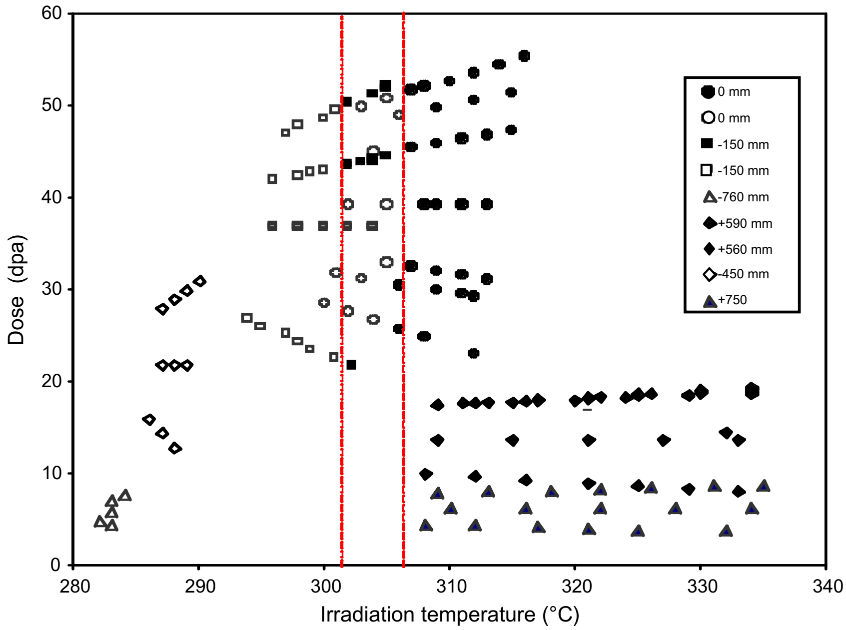

Fig. 1. Dose-temperature plot of occurrence of void formation of an austenitic alloy irradiated in the BN-350 reactor [4].

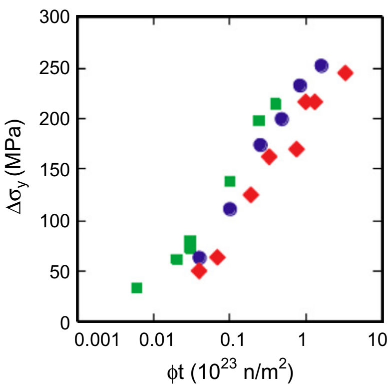

Fig. 2. Effect of dose rate (diamonds = high dose rate, circles = intermediate dose rate, squares = low dose rate) on hardening in a model RPV steel containing 0.4% Cu and 1.25% Ni [7].

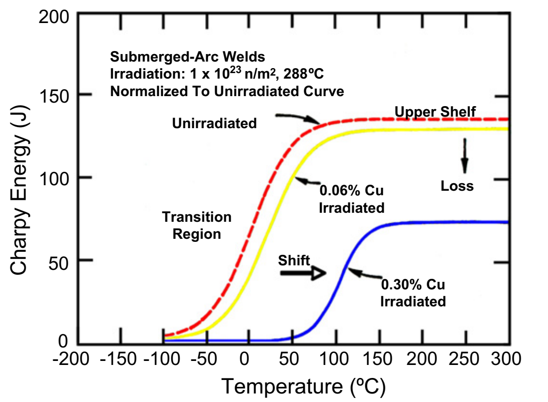

Fig. 3. Effect of 0.3% Cu on the DBTT and upper shelf energy for an RPV steel submerged-arc weld irradiated to 1×10^{23} n/m^2 at 288 °C [9].

cases, the results are very surprising. The effect of copper on embrittlement of RPV steel welds is well known. But a level as low as 0.3% can increase the transition temperature by over 100 °C and drop the upper shelf energy almost in half following irradiation to 1×10^{23} n/m^2 at 288 °C, Fig. 3 [9]. Cu-rich precipitate zones are responsible for about 80% of the transition temperature shift. Changes in the Si content of austenitic stainless steel from 0.5% to 1.0% is linked to severe IGSCC and enhanced crack growth rates [10]. The addition of less than 1 wt% Hf to 316 SS suppresses void formation in a high purity Fe–18Cr–12Ni alloy to beyond 50 dpa compared to less than 2 dpa in the undoped alloy, Fig. 4 [11].

2.4. Coupling of processes

Dose rate and temperature can combine to create severe effects. In EBR-II thimbles, the flux along the length of the core varies symmetrically from top to bottom, but the temperature peaks near the top, resulting in greater swelling at the bottom of the

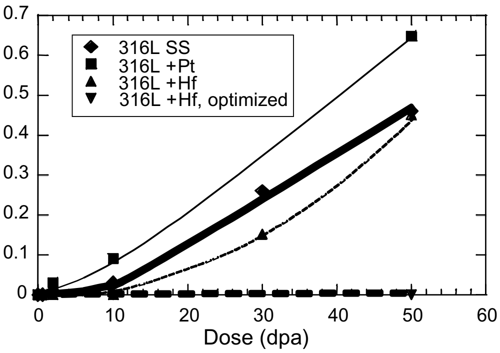

Fig. 4. Suppression of swelling to at least 50 dpa in 316SS doped with ∼1 wt% Hf after irradiation with Ni ions at 500 °C [11].

thimble compared to the top, but in locations that have experienced the exact same fluence, Fig. 5 [8]. The coupling of dose rate and temperature was exploited in the use of proton irradiation to emulate neutron irradiation effects. Nearly identical RIS profiles of Ni, Cr and Si were obtained by conducting proton irradiation at a dose rate ∼100× that for neutron irradiation and at a temperature 85 °C higher, Fig. 6 [12]. The advantage of coupling can be further realized by composition and microstructure modification. Addition of oversize solutes, grain boundary engineering and oxide dispersants can be combined to trap vacancies and reduce RIS, thus reducing IASCC, increasing creep resistance and enhancing strength [13].

2.5. Surface condition and welds

The importance of surface condition and welds in the performance of materials, especially in aggressive environments, is often underestimated. The Japanese BWR fleet was sidelined for several months due to SCC in core shrouds that was tracked to severe machining and grinding. Welds have continued to plague the industry and present a substantial challenge to maintaining component integrity. Reactor pressure vessel steel embrittlement, pipe cracking in BWRs, SCC in stainless steel core shrouds and cracking of alloy 182-alloy 600 welds in reactor vessel head penetrations in PWRs are some of the notable occurrences of problems with welds that have caused considerable problems in light water reactors over the years.

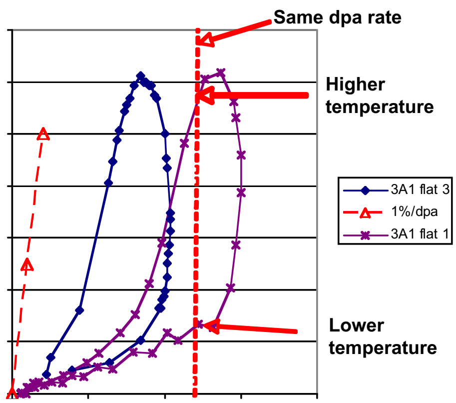

Fig. 5. Coupling of flux and temperature to produce a variation in swelling at locations where the fluence is the same [8].

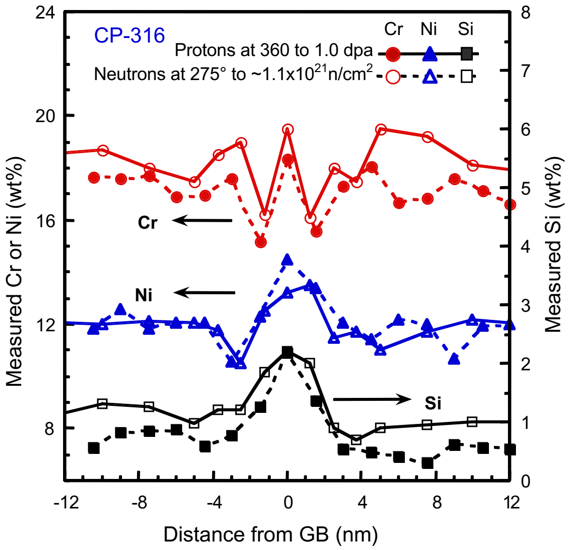

Fig. 6. Exploitation of the coupling between dose rate and temperature to achieve similar RIS profiles in proton-irradiated 316 SS irradiated at a dose rate ∼100× and a temperature ∼85 °C higher than neutron irradiation [12].

2.6. Gradients

While maximum values of key variables are used as the design point for systems, they are seldom uniform across the thickness or length of a component. Weld stresses in austenitic stainless steel piping in BWRs result in a residual stress distribution that varies through the thickness of the pipe. This variation is multiplied in the stress intensity factor variation at a crack tip, and must be accounted for in determining component lifetime. The reactor pressure vessel contains multiple variable gradients that can all impact its integrity [14]. As shown in Fig. 7, neutron fluence decreases from the inside to the outside of the vessel wall, the temperature increases

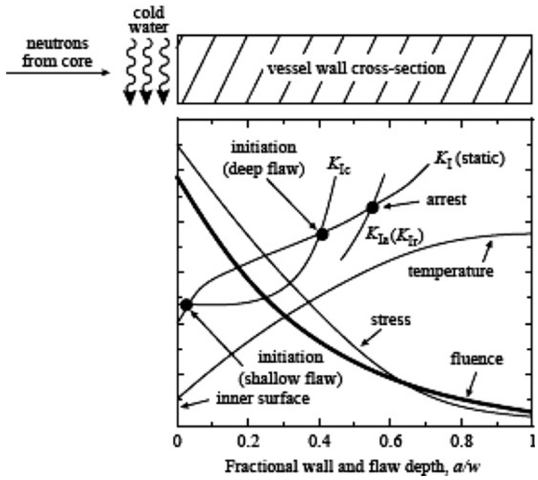

Fig. 7. Flux, temperature, stress and fracture toughness gradients in a reactor pressure vessel wall [14].

from the inside to the outside and the stress in the vessel peaks at the inside surface and decreases through wall. As a result of these gradients, the fracture toughness varies through wall in a complicated manner that must be understood in order to ensure safe operation of the vessel.

2.7. Incubation

Some of the biggest 'surprises' in material performance occur because of an incubation period that delays the emergence of a process. A classic example is void swelling in which composition, cold work and dose rate can push the onset of measurable

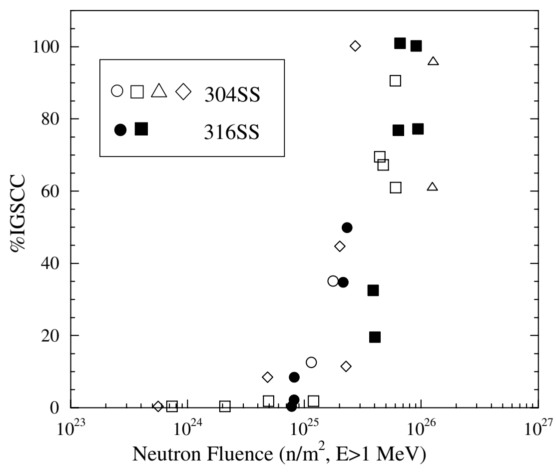

Fig. 8. Incubation dose for the emergence of IASCC in 304 and 316 SS in BWRs [16].

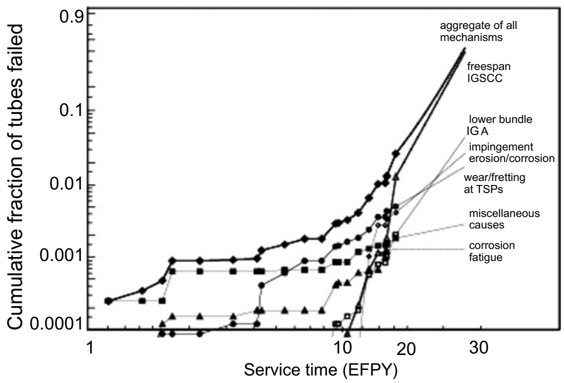

Fig. 9. Incubation in the degradation mechanisms in a PWR steam generator [17].

swelling from a few dpa to greater than 50 dpa [15]. IASCC in austenitic stainless steels has been observed to occur only after an incubation period of 1–3 dpa ($7–21 \times 10^{20}$ n/cm^2), Fig. 8 [16]. More extreme examples are cracking of alloy 182 welds or failure of steam generator tubes, Fig. 9, in which hot leg cracking required 6–10 years to emerge as a problem [17]! Unfortunately, once failures begin to occur, the rates can be extremely rapid. Although the emergence of hot leg cracking at the tubesheet and tube support plates in the Ringhals steam generators required almost a decade to appear as a problem, it took less than half that time for these processes to dominate all other failure modes combined.

2.8. Metallurgical variability

Material properties can vary substantially with metallurgical condition. Differences among alloy heats or batches can contribute to this variability in ways that are not fully understood. Yet heat-to-heat variability translates into variability in component performance that adds an uncertainty to the ability of plant operators to predict material performance. Fig. 10 shows the significance of heat-to-heat variabilities in the stress corrosion cracking of steam generator tubing [18]. Thirty six heats of tubing were used in a single steam generator in a single plant. Following 75 000 h of service, the performance in the tubes varied widely and the strongest variable explaining the variation of tube cracking throughout the SG was the heat. Note that while some heats had as much as one third of tubes affected, others had none. Yet all heats met the same specifications on composition and properties.

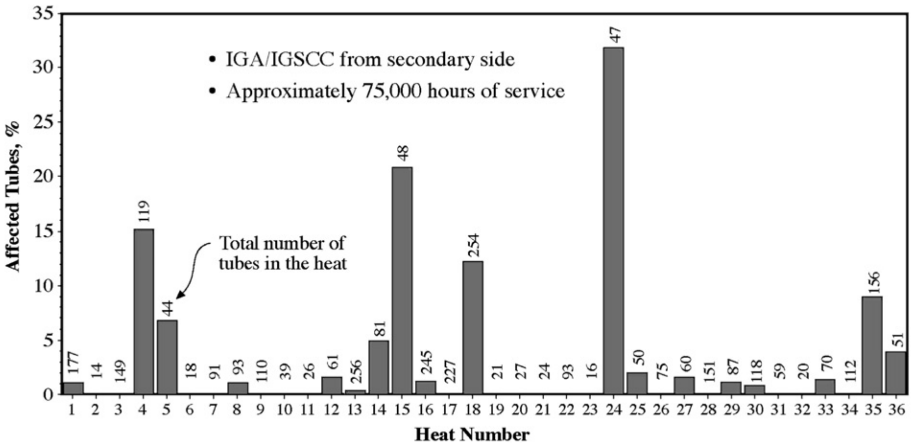

Fig. 10. Heat-to-heat variability in steam generator tube degradation after 75 000 h of service [18].

2.9. Environment (design)

One of the most important factors affecting component integrity, that is too often overlooked or discounted in the design process, is the environment. All power generation systems, fusion included, utilize a working fluid in converting the heat generated from the fuel to mechanical and then electrical energy. At the temperatures and pressures required for operation of nuclear sources, these fluids present aggressive conditions to the components they contact. Not surprisingly, corrosion continues to account for a majority of the capacity factor loss due to materials degradation in light water reactor systems. Steam generator tube cracking, vessel penetration weld cracking, baffle bolt cracking in PWRs and steam line cracking and core shroud cracking in BWRs, along with turbine disc cracking and cladding corrosion and SCC are only a few examples of environmentally-induced failures in light water reactors.

IASCC is a prime example of the prevalence of environmentally-induced degradation. IASCC is a form of intergranular stress corrosion cracking that occurs due to a combination of irradiation and an aggressive environment. It does not occur without the environment and may occur, but much more slowly, in the environment in the absence of irradiation. Yet it is a generic problem, affecting dozens of components in most austenitic alloys in all water reactor designs.

Steam generator tube cracking constitutes one of the most pervasive corrosion problems occurring in light water reactors. Its occurrence is a combination of a *susceptible alloy* and a *design* that promoted conditions conducive to corrosion. Staehle and Gorman [17] have identified some 25 distinct combinations of corrosion mode and location for degradation of steam generator tubes in steam generators with drilled-hole tube support plates.

As described earlier, advanced fission reactor concepts incorporate higher temperatures, higher dose rates and higher doses in an effort to boost efficiency and economy of power generation. The very high temperature gas reactor (VHTR), the lead-cooled fast reactor and the supercritical water reactor (SCWR) all present significant corrosion challenges due to aggressive environments and high temperatures. Heat exchangers proposed for the VHTR include the *printed circuit board* design that benefits from compactness and efficiency, but consists of very thin walls and numerous welds between layers. Both of these distinguishing features will present considerable challenges in maintaining material integrity at very high temperatures.

The SCWR will operate with supercritical water temperatures in the 600 °C range. Significant experience with supercritical fossil plants exists in the US and other countries, but the engineering is not directly transferable. For example, a ferritic–martensitic fire tube in a fossil plant may accumulate several hundred micrometers of oxide over 20 years of operation near 500 °C. For a 10 mm thick tube, this amount of oxidation is acceptable. But due to neutron economy, the SCWR fuel cladding thickness is closer to 0.6 mm and the water rod design calls for 0.4 mm thick walls. At the same corrosion rate as in a fossil plant, the lifetime would be limited to the order of a year at best, without accounting for the effect of irradiation on corrosion and SCC.

Because the bulk of the power generation plant is similar for all systems, and because the environment for materials in advanced fission reactor systems is moving closer to that in fusion systems, fusion will face the same problems as those just discussed. As concepts evolve into designs, the fusion community should look to the LWR industry and the advanced fission reactor program as a repository of a rich history of material performance data, and for guidance in dealing with generic issues involving materials performance in power generation systems. However, material performance is only part of the issue that the fusion community must confront. The other is component failure and failure prediction.

3. The importance of failure (or performance) prediction

The operation of materials in high temperature, aggressive environments *will* lead to degradation, and ultimately, failure. The capability to predict and track the approach to failure has enormous economic and performance consequences. The development of sophisticated performance prediction capability is required in order to provide defensible arguments to regulators for the continued safe operation of a system containing flaws. The inability to do so will result in the application of unnecessarily conservative assumptions that will force premature shutdown and component replacement, all of which can be extremely costly.

Over the years, the LWR industry has evolved from *deterministic* failure prediction to *probabilistic* prediction methodology. Historically, laboratory and commercial experience were used to develop a phenomenological model for the failure of a component. However, such deterministic approaches often have poor accuracy due to the multitude of variables affecting the outcome, a poor knowledge of the failure mechanism and uncertainty in many of the materials parameters and dependencies. As a result, probabilistic failure prediction has gained importance in predicting the behavior of complex systems.

An early example of probabilistic failure prediction is the failure of fuel rods in light water reactors. Using fuel rod pre-characterization parameters, reactor operating history, thermo-mechanical fuel behavior models and deterministic failure models as input, Christensen [19] used pattern recognition methodology to predict fuel failures in an operating reactor. The prediction called for 6 ± 3 of the 217 fuel assemblies in the Maine Yankee reactor cycle 4 (September 1978–January 1980) to contain at least one leaking rod. Sipping results in January 1980 revealed that there were 9 assemblies with at least one leaking rod. Interestingly, the contribution of the deterministic model to this extremely accurate result was so small as to be inconsequential. This early example highlighted the potential for statistical modeling of failure processes in nuclear systems.

More current examples include recent work by Staehle [20] on the development of statistical models for steam generator tube failure. Staehle developed a relation between the dependent variable, e.g., depth of SCC penetration, and the independent variables such as temperature, stress, and pH then determined the dependence of the statistical model parameters on the independent variables. The objective is to model the statistical parameters according to the dependencies of the primary independent variables in order to better model the crack penetration depth (and hence, the failure distribution) in time.

Probabilistic fracture mechanics is also being applied to degradation of reactor pressure vessels. A model is constructed for the stress intensity factor as influenced by design, physical properties of the component, flaw data and thermal/hydraulic system performance. Another model is developed for the irradiation temperature shift that depends on alloy chemistry, fluence, temperature, etc. The models are combined with a fracture mechanics model to provide the probability of crack initiation and the probability of thru-wall cracking, Fig. 11.

Prediction of materials degradation and failure are important for maintaining the integrity of the component and the safe operation of the plant. But the light water reactor industry has gone one step further in materials management in reactor systems – proactive management of materials.

4. Proactive materials management

Two separate but similar initiatives have been established to actively seek out and identify the potential weak-links in materials performance and address the weaknesses *before* they emerge in plant operation. This proactive approach is designed to pre-empt problems before they occur. One such program is the Industry Initiative on Management of Materials Issues that was established in 2003 by the chief nuclear officers of the nation's utilities under the auspices of the Nuclear Energy Institute

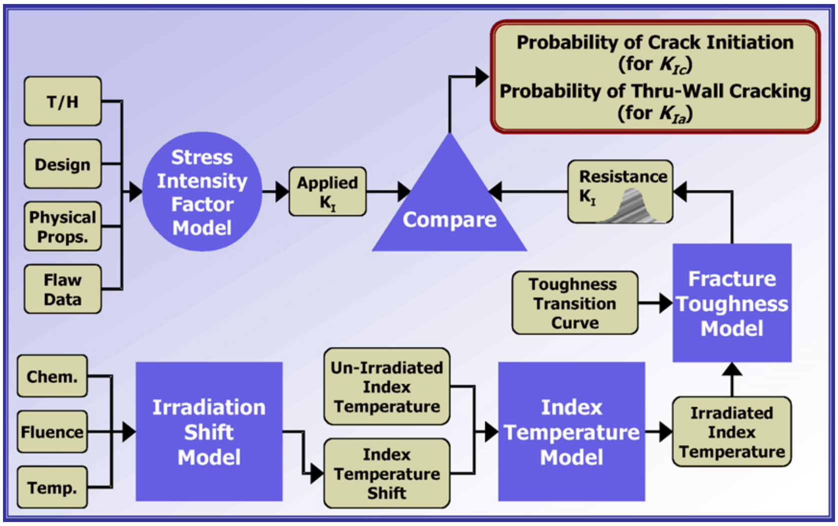

Fig. 11. Probabilistic approach to failure of a reactor pressure vessel steel. (Courtesy S. Rosinsky, EPRI).

and the Electric Power Research Institute [21]. The purpose of the initiative is to assure the safe, reliable and efficient operation of US nuclear power plants by providing for:

- consistent materials degradation issue management processes across the industry,
- prioritization of materials degradation issues, and
- proactive, integrated and coordinated approaches to issue resolution.

The approach utilizes two tools; degradation matrix tables and issue management tables. The degradation matrix tables seek to identify the materials used for major passive components and systems, identify the most probable degradation modes of each component of each system of a particular reactor type, and develop the tools to manage the potential threats to system integrity. The issue management table focuses on identifying specific components, materials of construction, failure mechanisms and locations, consequences of failure and mitigation/repair/replacement options, and goes so far as to identify knowledge gaps and prioritize work to resolve the gaps.

The NRC is in the process of developing a Proactive Materials Degradation Assessment program with the objectives of identifying materials and locations where degradation can reasonably be expected in the future, and developing and implementing an international cooperative research program for the components and degradation of interest that will review, evaluate and develop remedies as needed.

Both programs have in common an exhaustive and thorough examination of every system, sub-system and component in the plant. While time consuming and expensive, only by combing through each and every component in every sub-system in every system, can potential problems be identified and addressed. The fusion community can benefit from these pioneering efforts by implementing proactive programs as specific designs begin to take form.

5. Summary

Materials degradation has played a large role in the loss of capacity factor in the fission reactor industry. Degradation is costly and can limit the competitiveness of the power system. But it is also inevitable as corrosion, for example, is nothing more than the return of a metal to its thermodynamically stable state, and engineered metal-based systems are the antithesis of this process.

While material behavior in reactor systems is influenced by many parameters and processes, its sensitivity to some parameters or combinations of parameters is notable. In particular, temperature, dose rate, composition/microstructure, surface condition and the coupling of multiple parameters have played key roles in fission materials performance. Processes such as incubation time for the occurrence of a degradation mode and gradients in thick sections have also driven materials performance. The variability in metallurgical condition adds a confounding parameter to materials performance prediction. Environmental effects have played a major role in LWR materials performance and will likely be a key factor in fusion systems as well.

When concepts evolve into designs, the assurance of safe operation of a reactor system will depend on the development of failure prediction models. These models have evolved from purely deterministic to statistical or probabilistic, in an effort to accommodate the numerous variables affecting materials behavior and poorly defined degradation mechanisms.

In an effort to move beyond a purely reactive mode of materials performance, proactive materials management programs are being developed to identify weak-links in components and systems and to develop the knowledge and tools in a pre-emptive fashion. The identification of key parameters and processes that affect materials performance in reactor systems, the evolution of prediction methodology for materials performance and failure, and the advent of proactive materials management programs provides a rich toolbox for the efficient development of future fusion reactor systems. The fusion community has much to gain by drawing on the efforts of the fission reactor industry to address materials degradation issues, develop models for their occurrence and begin to establish proactive materials management plans to pre-empt problems that can be foreseen.

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