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Investigation of magnetic signatures and microstructures for heat-treated ferritic/martensitic HT-9 alloy

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Abstract

There is increased interest in improved methods for in situ non-destructive interrogation of materials for nuclear reactors in order to ensure reactor safety and quantify material degradation (particularly embrittlement) prior to failure. Therefore, a prototypical ferritic/martensitic alloy, HT-9, of interest to the nuclear materials community was investigated to assess microstructure effects on micromagnetics measurements (Barkhausen noise emission, magnetic hysteresis measurements, and first order reversal curve analysis) for samples undergoing three different heat treatments. Microstructural and physical measurements consisted of high precision density, resonant ultrasound elastic constant, Vickers microhardness, grain size, and texture determination. These were varied in the HT-9 alloy samples and related to various magnetic signatures. In parallel, a mesoscale microstructure model was created for α-iron and the effects of polycrystallinity and the demagnetization factor were explored. It was observed that Barkhausen noise emission decreased with increasing hardness and decreasing grain size (lath spacing), while coercivity increased. The results are discussed in terms of the use of magnetic signatures for the non-destructive interrogation of radiation damage and other microstructural changes in ferritic/martensitic alloys.

Keywords: Ferritic steels; Magnetic properties; Hardness; Ferromagnetic; Non-destructive evaluation

1. Introduction

The existing US nuclear fleet has a remarkable safety and performance record, and today most of these reactors have either received or applied for an extension to their operating license, from the initial 40 years to a period of 60 years. In its Research and Development Roadmap – Report to Congress dated April 2010 (www.ne.doe.gov) the DOE Office of Nuclear Energy identified the development of technologies and other solutions to improve the reliability, sustain the safety, and extend the life of current reactors as its top priority. Simultaneously, research into advanced reactor designs with passive safety features continues, with many of these reactors expected to operate at higher temperatures than light-water-cooled reactors. In both light-water-cooled and advanced reactors, maintaining the structural integrity of passive safety components (such as the reactor pressure vessel or reactor coolant system piping) will be necessary to ensuring safe long-term operations. In order to achieve this objective, non-destructive structural health or condition monitoring techniques must be integrated with plant operations to quantify the "state of health" of structural materials. In addition, new reactors have the option of being designed for better inspectability in order to enhance safety.

In addition to this, the Nuclear Regulatory Commission stated

> "Material characterization using NDE [non-destructive evaluation] is being developed to produce more accurate, in situ evaluation of the structural integrity of degraded components and radiation damage. This is

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> promising work because many NDE methods are sufficiently sensitive to the presence of residual stress, while also being sensitive to microstructural material variations that usually accompany residual stresses and aging" (NUREG-1925, Rev. 1, December 2010, US Nuclear Regulatory Commission Office of Nuclear Regulatory Research, Washington, DC, available at www.nrc.gov).

The goal of in situ evaluation is lofty but appears to offer possibilities with regard to improved reactor safety by being able to assess the state of a material in some manner that can be related to damage and degradation. This research discusses the use of advanced magnetic signatures and methods that could potentially be used to monitor materials hardening due to radiation damage accumulation during reactor operation.

As far back as 1953 the use of magnetic Barkhausen noise and coercive force measurements were used to interrogate steels and other alloys to assess the microstructural state, particularly with regard to composition and strengthening precipitates [1–3]. Early work on the effects of irradiation and radiation damage on the magnetics and micromagnetics of materials demonstrated that the majority of materials were sensitive in some degree to changes in microstructure due to irradiation [4], in one case demonstrating a linear response to neutron dose [5]. More recently, concerns regarding embrittlement of reactor pressure vessel (RPV) steels encouraged research into nondestructive evaluation (NDE) methods to determine mechanical property changes due to neutron irradiation, and magnetic measurements were explored with some success. One of the first investigations used magnetic acoustic emission (MAE) and magnetic Barkhausen noise (MBN) to investigate radiation-induced changes in A302B plate and A533B weld steels [6]. The analysis concluded that MAE waveform analysis was a promising NDE technique for monitoring the microscopic changes in steel components subjected to neutron irradiation. Other magnetic measurements, including remanence, coercivity, and permeability were also used to study radiation damage in these same steels. Magnetic remanence and maximum permeability were found to be highly sensitive to neutron dose [7]. It was hypothesized that atomic disorder due to neutron irradiation caused large-scale magnetic domains to break down, while the resulting smaller domains more easily rotated to form closed flux loops, thus decreasing remanence. The magnetic signals were more sensitive to these microstructural changes than mechanical properties, such as microhardness, in this study [8].

Others realized that micromagnetics could be used for NDE of both nuclear pressure vessel steels [9–14] and power plant steels [15]. Consequently, these steels were soon being routinely investigated using these methods and correlations were developed for hardening and aging. The use of Barkhausen signals to monitor stress relief of welds was also pioneered during this time [16], as was thermal recovery monitoring of RPV steels [13]. At the same time some innovations in these magnetic methods were also being explored, such as the Pulse MAE method [17], which demonstrated that signatures increased monotonically with neutron dose. In another study a SQUID sensor was used to demonstrate that magnetic coercivity could be used as a signature for radiation hardening with good sensitivity [18]. Giant magneto-impedance measurements were also shown to reveal similar signatures [19]. More general uses for micromagnetics measurements were also emerging, such as monitoring tensile and fatigue strain as a measure of degradation [20]. Strong correlations are realized between neutron irradiation mechanical property changes and micromagnetic signatures when multiple methods are combined using statistical techniques [21]. More recently these methods have become almost routine for RPV materials in terms of monitoring hardening and recovery [22–24]. However, such micromagnetic methods cannot address all such embrittlement problems, such as phosphorus segregation when no hardening is involved [25].

The trends with regards to hardening and neutron irradiation effects are clear, however, and magnetic methods that are sensitive to domain wall pinning are suitable for NDE of magnetic materials. Regardless of the alloy, MBN almost always decreased with irradiation [26] due to domain wall pinning, and typically coercivity and permeability increased, though not always [13,27]. Recent results [28–30] suggest that the variation in coercivity with fluence, or yield strength and/or hardening, is a function of the alloy composition and initial microstructure. Irradiation processes that produce point defects and small defect clusters can be effectively probed as a function of dose for small doses using these methods, and linear responses are found initially. Embrittlement thresholds can be determined using statistical regression methods and calibrated samples. However, these methods cannot distinguish between hardening events and researchers often suggest that additional work is required to sort out complex effects.

With regard to HT-9 and other ferritic alloys, several studies have found that proton irradiation can modify the internal magnetic properties via Cr depletion of the Fe–Cr matrix rather than via radiation hardening [31,32]. There is no direct data, to the best of our knowledge, that relates HT-9 radiation hardening to other magnetic property changes, such as MBN or hysteresis/coercivity. However, in general radiation hardening of Fe [33] and model ferritic alloys [34] can be directly correlated to MBN, which indicates that unirradiated HT-9 can be used as a surrogate material for irradiated HT-9 using conventional hardening methods. Conventional martensitic hardening is suggested, therefore, to be a surrogate for radiation hardening by modifying the magnetic response of HT-9 due to domain wall pinning. In turn, domain pinning changes should be observable using MBN and other hysteretic methods that we wish to develop for NDE uses. It must be noted, though, that other changes due to irradiation, such as Cr depletion and elemental segregation, are very difficult to