X-RAY DIFFRACTION ANALYSIS ON PRECIPITATES OF 11J IRRADIATED RAFS—H. Tanigawa (Japan Atomic Energy Research Institute), H. Sakasegawa (Kyoto University), E. A. Payzant, S. J. Zinkle, and R. L. Klueh (Oak Ridge National Laboratory), and A. Kohyama (Kyoto University)

OBJECTIVE

The objective of this work is to analyze the precipitation behavior of irradiated reduced activation ferritic/martensitic steels (RAFs) by X-ray diffraction (XRD) analysis.

SUMMARY

XRD analyses were performed on the extraction residue of HFIR 11J-irradiated RAFs to investigate the overall precipitate character. Un-irradiated and aged specimens were examined as well. Results suggested that the distinctive peaks of $M_{23}C_6$ (M; Cr, Fe, W) were observed on all specimens. Peaks possibly related to MX (M;Ta,Ti,V : X ; C, N) were observed on the specimens extracted from un-irradiated JLF-1 and ORNL9Cr, but those peaks were not observed on irradiated specimens.

PROGRESS AND STATUS

Introduction

XRD analysis on an extraction residue specimen is the commonly used technique to analyze the precipitate characteristics on the R&D of steels in an irradiation-free condition. This technique has not been used for irradiated steels because of the practical limitation on making and handling the neutron -irradiated specimens. Transmission electron microscope (TEM) observation on thin film specimens or on extraction replica specimens is the most commonly used technique for precipitate analysis [1,2]. The conclusion obtained from TEM thin film and extraction replica specimens is that the results usually represent a very local microstructure, and it requires a lot of work to obtain a certain level of statistical reliability on the results. In order to cover the above problem, the authors decided to develop the extraction residue procedure to perform XRD analysis on irradiated steels.

In this study, XRD analyses were performed on the extraction residue of HFIR 11J-irradiated RAFs, which showed a variety of brittleness-hardness combination, to investigate the overall precipitate character. The un-irradiated and aged RAFs were also examined for comparison.

Experimental

The material used was IEA-modified F82H (F82H-IEA). Base metal with two heat treatment variations (standard IEA heat treatment and another heat treatment designated HT2) was irradiated. ORNL9Cr-2WVTa (ORNL9Cr), JLF-1 HFIR heat (JLF-1) and 2% natural Ni-doped F82H (F82H+2Ni) were also irradiated for comparison. Details of the chemical compositions and the heat treatments are shown in other reports [3,4]. Irradiation was performed in the Oak Ridge National Laboratory (ORNL) High Flux Isotope Reactor (HFIR) up to 5 dpa at 573K in the removable beryllium (RB) position. Specimens selected to be used were the 1/3-size Charpy specimens. Unirradiated (normalized-and-tempered) specimens of all materials and aged specimens of ORNL9Cr (873K for 10k h) were also used for comparison. The extraction residue from these specimens was collected on a filter with 200 nm pore size [5].

Samples were analyzed using PANalytical X'Pert Pro MPD x-ray diffractometer with a Ni-filtered Cu K-alpha radiation source at 45kV and 40mA. The diffractometer was configured with an incident beam multilayer mirror optic and diffracted beam parallel plate collimator, which yield parallel beam optics and thereby minimize sample surface displacement errors (mainly associated with sample mounting). Axial divergence limiting Soller slits were used on both the incident and diffracted beam, and the detector was a gas proportional counter. The fine powders were examined as-deposited on the filter materials and radiological safety requirements necessitated covering the powders with a thin film of Mylar (6.35μm thick) to prevent possible contamination of the system (Fig. 1). Consequently the relatively weak sample diffraction data was combined with diffraction from the Mylar and the filter. The diffraction data were analyzed using Jade version 6.5 software [Materials Data Inc., Livermore, CA].

Results and Discussion

Fig. 2 shows the raw peaks obtained from irradiated F82H-IEA, Mylar, background, filter, and the processed peaks of irradiated F82H-IEA. Background peak was obtained by extremely lowered X-ray energy. It is clear that there is no effect on the result from radiation, though the sample has an activity about 20mR/hr at contact. Mylar has a large wide peak at 18 degrees, but this does not affect the results as we used the peaks above 30 degrees for analyses. The filter has several distinctive peaks above 30 degrees. The peaks are matched to manganese phosphorus nitride (Mn_2PN_3). These peaks were removed from the original scan by subtracting the appropriately scaled diffraction pattern of the filter itself.

Fig. 3 shows the results of peaks from the extracted residue of unirradiated and irradiated RAFs. It revealed that the majority of precipitates are $M_{23}C_6$ type (denoted with diamond mark) for all as-prepared and irradiated steels. The 3 distinctive peaks (denoted with triangle) are observed on as-prepared JLF-1 and ORNL9Cr, and those peaks are not detected on irradiated specimens. There is no exact match pattern for those peaks, but the best match is for the TaC peaks. This supposition is quite reasonable as a high number density of Ta-rich precipitates is observed on these steels [6]. The reason for this peak offsets would be that TaC is usually present as MX precipitate (M; Ta, V, Ti : X; C, N). This disappearance of MX peaks is a very distinctive feature of the irradiation effect, because MX precipitate itself is thermodynamically stable at 573K.

The stability of MX precipitate in the unirradiated condition was suggested from the peaks of aged ORNL9Cr (Fig. 4), as MX peaks are still obvious even after 873K for 10k h aging. It should be noted that the peaks of aged ORNL9Cr also show that they have Laves phase (Fe_2W) in their residue, and this is very common in this type of steel.

The other feature obtained from Fig. 3 is the decrease of the $M_{23}C_6$ peaks of F82H-2Ni after irradiation. The weight of the residue itself was increased, as shown in other reports [6], so this change could be interpreted as

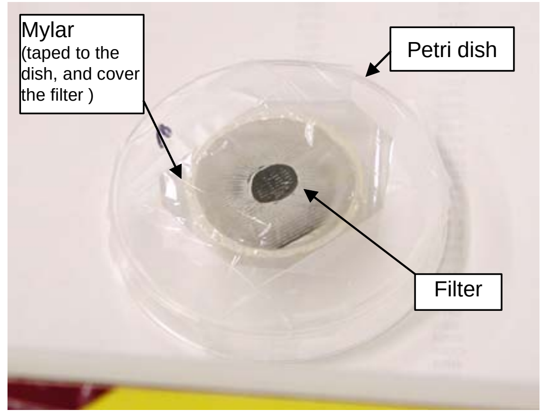

Fig. 1. Prepared status of the extraction residue sample of irradiated steels for XRD analyses.

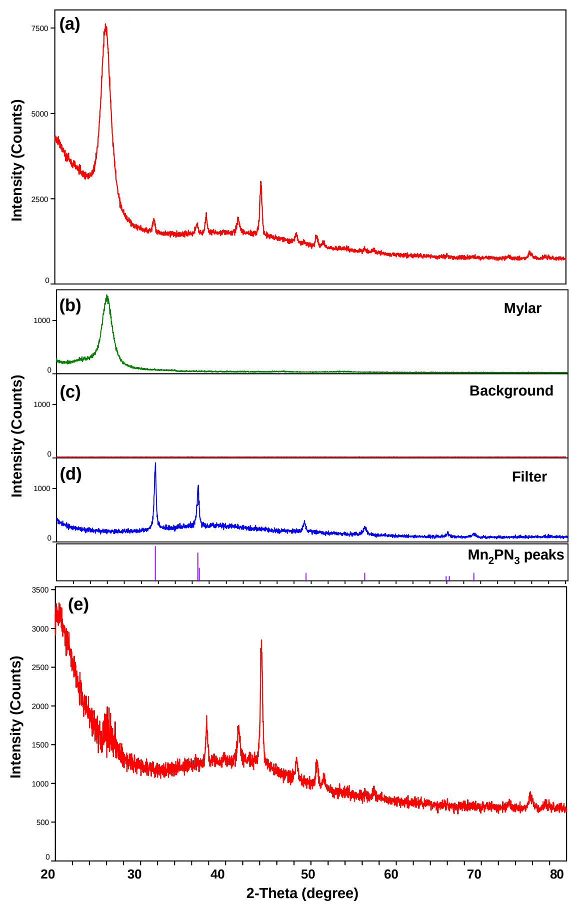

Fig. 2. Raw XRD peaks of (a) irradiated F82H-IEA, (b) Mylar, (c) background, (d) filter, with (e) processed peaks of (a).

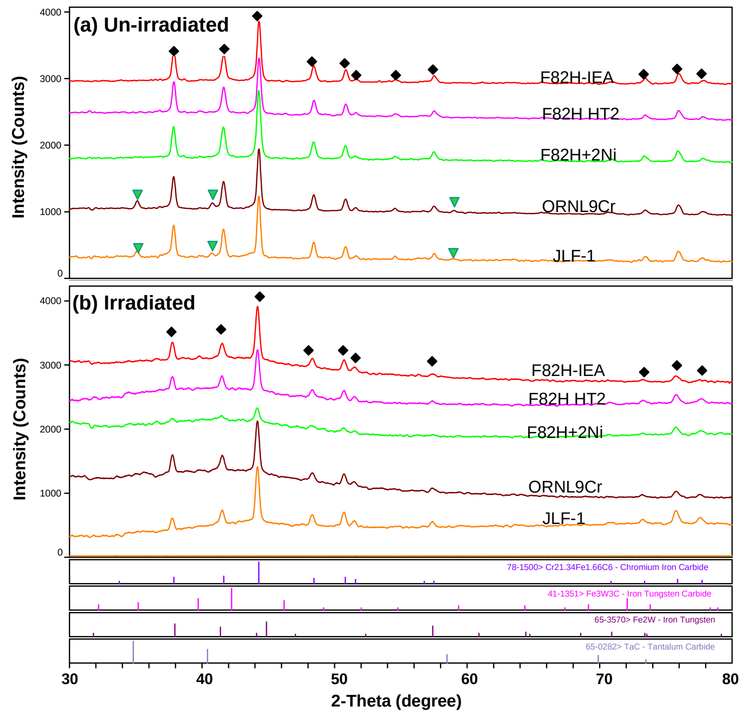

Fig. 3. XRD peaks of extracted residue from un-irradiated and irradiated RAFs. Peaks marked with diamond correspond to the peaks from $M_{23}C_6$, and with triangles correspond to peaks from MX (TaC).

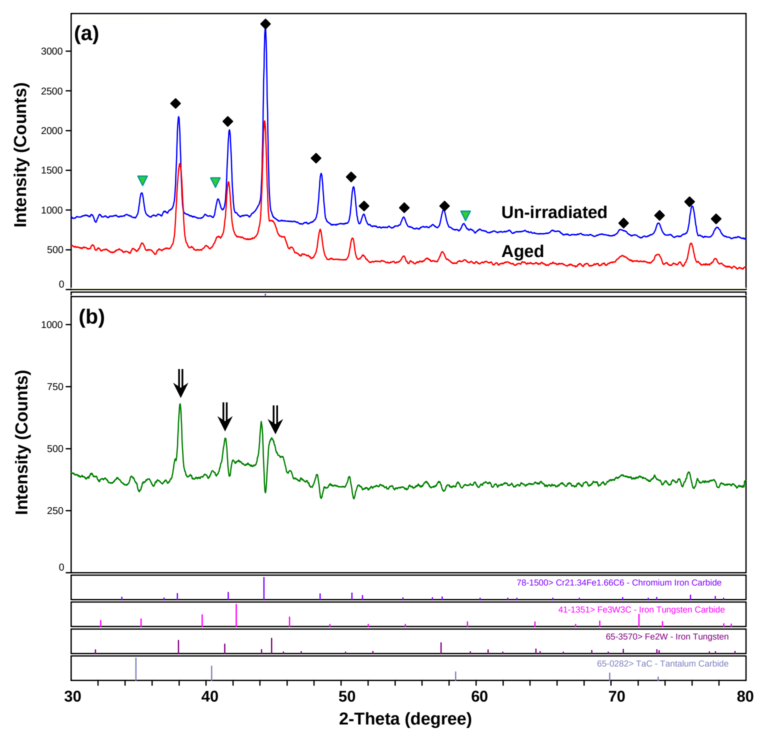

Fig. 4. XRD peaks of (a) un-irradiated and aged ORNL9Cr and (b) peaks obtained by subtracting the peaks of un-irradiated ORNL9Cr from peaks in (a). Black arrows suggest the peaks correspond to Laves (Fe_2W) phase.

the average size decrease of $M_{23}C_6$. It also should e noted that the peaks of all irradiated specimens tended to have broad and shallow peaks with the median of about 42 degrees. This might be related to the peaks from fine particles of M_6C (Fe_3W_3C), as its highest peak located at 42 degrees. This suggests the possibility that fine M_6C (M; Fe, W, Cr: nominally Fe_3W_3C) may be present. Further investigation on these changes will be performed by detailed TEM observation.

SUMMARY AND CONCLUSIONS

XRD analyses were performed on the extraction residue of HFIR 11J-irradiated RAFs to investigate the overall precipitate character. Unirradiated and aged specimens were examined as well. The following is a summary of the important observations and conclusions:

(1) The most distinctive peaks are of $M_{23}C_6$ (M; Cr, Fe, W) on all specimens.
(2) Peaks possibly related to MX precipitates were observed on the residue from un-irradiated JLF-1 and ORNL9Cr, but those peaks were not observed on irradiated specimens of those steels.
(3) The above peaks are observed on 873K 10k h aged ORNL9Cr.
(4) This disappearance of MX peaks is probably the effect of irradiation.
(5) The decrease of peak intensity after irradiation was observed on Ni-doped steel, and the relation to the average particle size decrease was suggested.
(6) The possibility of the presence of fine M_6C in the irradiated steels was suggested.

Acknowledgements

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