HEAVY ION IRRADIATION ON U-Mo/Al SYSTEMS

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Abstract

High density U-Mo/Al based nuclear fuels are considered as the most prospective candidates for converting high flux research and test reactors towards operation with lower enriched uranium fuels. During in-pile irradiation, the growth of an amorphous interaction layer (IL) at the U-Mo/Al interfaces limits the overall fuel performance. Such an amorphous IL can as well be generated by the out-of-pile technique of heavy ion irradiation performed at the MLL Tandem accelerator. Both XRD and TEM measurements evidenced the strong accordance between results obtained in-pile and out-of-pile. Therefore, heavy ion irradiation allows efficient and economic pre-qualification tests of future U-Mo/Al fuel plates. Among these qualification tests is the application of a diffusion barrier element “X” between the U-Mo and the Al to prevent IL growth. Several heavy ion irradiation experiments were carried out on such U-Mo/X/Al systems and post-irradiation examinations indicated promising barrier elements.

INTRODUCTION

Among the high uranium density fuels for conversion of research and test reactors, the alloy of uranium-molybdenum (U-Mo) with 7 to 10wt% Mo content is considered as the most promising option to achieve this goal [1]. An U-Mo based nuclear fuel typically consists of U-Mo powder particles embedded in an Al matrix, although monolithic U-Mo/Al layer systems are considered as well [2]. During in-pile irradiation tests, the desired fuel performance could not be achieved as an interaction layer (IL) has grown between the U-Mo fuel and the surrounding Al matrix. This IL has been found to be amorphous at irradiation temperatures below 200°C and has a main elementary composition of UAl_x [3, 4]. Furthermore, these ILs cannot retain fission gases which accumulate inside large bubbles leading to an exponential fuel plate swelling and eventually to a fuel plate integrity loss. Therefore, one major objective of U-Mo/Al fuel development is the suppression of IL formation. Among the envisaged solutions is the addition of another element to the matrix or the protective U-Mo particle coating by a diffusion barrier element [5, 6, 7]. Numerous in-pile irradiation tests would be required as part of the fuel optimization process. Therefore, alternative out-of-pile qualification tests are strongly encouraged. Thermal annealing has been widely applied to generate an IL. However, these ILs mainly consist of crystalline phases and even ternary compounds, e.g. $U_xMo_yAl_z$ [8, 9]. Both properties are not observed after in-pile irradiation.

At the MLL Tandem accelerator, another approach for out-of-pile studies was performed by FRM II's fuel development group (“Arbeitsgruppe Hochdichte Brennstoffe”) in collaboration with the French “Commissariat a l'energie atomique et aux energies alternatives” (CEA). Indeed, it has been demonstrated that an IL has grown in U-Mo/Al systems after irradiation with ^{127}Iodine at 80 MeV, which is a typical uranium fission product and fission energy [10, 11]. Post-irradiation examinations of the IL showed the same elementary composition (e.g. UAl_x) as inside those ILs obtained after in-pile irradiation [12]. Based on this good agreement, several studies were performed to qualify rate-selected fuel optimization options.

RECENT STUDIES

Two major enhancements of the irradiation setup at the MLL were realized during the past two years. First, the complex and random structure of the actual dispersed fuel samples was replaced by a well defined sample geometry which was obtained by PVD sputter coating of U-Mo on an Al substrate. These obtained layer systems have a very well defined global U-Mo/Al interface, and therefore allow for a much deeper insight into the IL growth mechanism. Second, a new experimental setup was constructed which allows a very stable long time irradiation with only a minor temperature deviation of ±1°C. The final fluence of $1 \times 10^{17}\ \frac{ions}{cm^2}$ was achieved with a particle flux of $1.2 \times 10^{12}\ \frac{ions}{s\bullet cm^2}$, notably lower than in preceding experiments ($\approx 1.5 \times 10^{12}\ \frac{ions}{s\bullet cm^2}$).

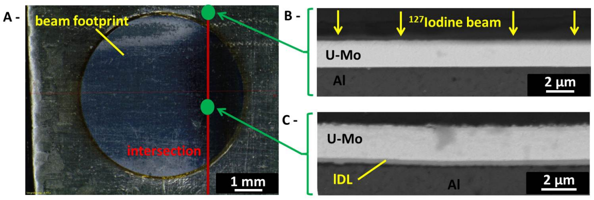

Figure 1: (A-) Image of an irradiated U-Mo surface. The red line indicates the intersection for SEM cross-section examination. Two representative spots are shown in (B-) and (C-): (B-) Areas not exposed to the Iodine beam show no IL growth, while those regions affected show an according interaction (C-).

An IL with a very homogenous layer thickness has grown in these monolithic layer systems (see fig. 1) [13]. Both post irradiation experiments, TEM and nano-XRD, have demonstrated that the generated IL has amorphous compounds. This striking feature was not observed in the samples irradiated at higher flux and at higher temperature [11, 12]. Therefore, by choosing a well defined irradiation temperature, particle flux and sample geometry, heavy ion irradiation is an ideal tool to study the IL growth dependence on these irradiation parameters.

Another application is the testing of the afore-mentioned diffusion barrier elements for avoiding IL growth at all. Several possible barrier elements were studied, among them Ti, Nb and Zr. All these three elements show excellent results regarding IL negation. Several of these were performed within an international collaboration, including the Belgian SCK-CEN [14] and the Korean KEARI institutes [15]. In both experiments it could be demonstrated that U-Mo particle areas not protected by coatings are exposed to a high level of U-Al interaction. Identical observations after in-pile irradiation of these fuels were made, giving evidence for the strong comparability between heavy ion and in-pile irradiation. Due to these promising results, the international collaborations considering heavy ion irradiation at the MLL are continued.

OUTLOOK

Several future applications for Iodine irradiation on U-Mo/Al systems are envisaged:

- First, fission gas behaviour inside the IL has a strong contribution to the fuel's in-pile performance. By combining Iodine irradiation to generate ILs and successive Kr implantation into those ILs, out-of-pile-studying of inert gas behaviour would be possible as well. A first approach was already made when Kr was implanted at the GANIL accelerator in Caen, France. First SEM observations indicate that the implanted Kr particles show the same behaviour as fission gases inside in-pile irradiated fuel, e.g. accumulation in micrometer-sized bubbles. Further implantation experiments with varying Kr flux, fluence and irradiation temperature are encouraged to provide a global overview over Kr and IL behaviour inside the layer systems. Therefore, initial IL generation by heavy ion irradiation performed at the MLL is mandatory.

- Especially fuel optimization by particle coating processes and/or fuel matrix modification is of high interest. Further numerous qualification tests would be necessary. An already ongoing international collaboration in the fuel development sector (HERACLES group) is planning measurements in the heavy ion irradiation field. An efficient selection of prospective fuel candidates is desired before actual time-consuming in-pile irradiation tests are launched.

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