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DEVELOPMENTS ON RESEARCH REACTOR DISPERSED FUELS IN THE WORLD - PERSPECTIVES AND TENDENCIES

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ABSTRACT

The purpose of this paper is to present the development status in the world related to dispersed fuel elements for research reactors. The present paper provides a review on nuclear fuels for thermal research and test reactors. The review covers a historical background, some recent technological advances, fabrication procedures and problems, and the new development efforts, with emphasis on RERTR program activities that have had an important role in the dispersed fuel development worldwide scenario. Since the beginning of this program in 1978 there has been a significant development in the dispersed-type fuel element field. In 1988, the U. S. Nuclear Regulatory Commission approved fuels up to 4.8 g U/cm^3, after a development period of about 10 years, allowing the conversion of the vast majority of research reactors from the use of high to low enriched uranium. CERCA has reported the fabrication capability of silicide fuels up to 6 gU/cm^3 and some developments involving nitride uranium fules. The conversion of the IEA-R1 research reactor from HEU to LEU was decided in 1981 and must be completed in 1997, when all core must be constituted by U_3O_8-Al fuel elements produced at IPEN. Simultaneously, there is an ongoing program to increase the IEA-R1 power from 2 to 5 MW. This review is an attempt to present the state-of-the-art of the dispersion fuels for research and test reactors and to discuss some major problems involved in the higher uranium loading fuel fabrication, taking into account the IEA-R1 updating program and the IPEN future fuel development possibilities.

I. INTRODUCTION

The current plate-type aluminum-dispersed fuel technology has an important role in the reactor performance and in the reactor operational costs. The main characteristics of fuel plates should be an excellent irradiation performance, with negligible swelling at high burnups (up to 90 atoms %), and cladding integrity. In order to keep the reactor performance at a reasonable cost, taking into account the constraints imposed by international policies related to uranium trading, technological activities have been conduced to develop new fuel compounds and fuel plates with higher uranium loading to compensate the enrichment reduction [1].

Besides other important parameters, the amount of ^{235}U in the core of a research reactor controls the period without need new fuel elements. The uranium loading can be increased without changes in the fuel plate dimensions by higher uranium densities and/or reducing the clad thickness. Of course, there are limits imposed by the technology availaible and safe concerns. The aim at increasing the uranium density is: to improve the performance; to reduce the fuel element consumption (by reducing the cost/year of manufacturing and by reducing the number of irradiated elements, with impact in the end of the cycle management); the conversion of the high performance reactors [2].

Metallic uranium would be a very interesting option to increase the uranium loading in fuel plates. Nevertheless, the unstable uranium α phase swells under irradiation. The current most advanced tested fuel is the U_3Si_2 dispersed in aluminum with a uranium density upper limit of about 5 or (optimistically) 6 g/cm^3[3]. Nowadays, non-proliferation policies are pursuing the goal of high enrichment uranium replacement for low enrichment for all research reactor types, even for those of high performance. Although a fuel with a uranium density of 4.8 g/cm3 is sufficient to convert about 90 % of the research reactors which used HEU (from U.S. origin), conversion of the remanescent reactors requires fuels having higher U density [4]. Besides the development of new acceptable compounds. and the related fabrication technology, the good operational performance has to be tested under irradiation conditions in order to demonstrate the new fuel concept. In the following sections a review of the main problems involved in the research reactor fuel element fabrication and development is presented.

II. BACKGROUND ON RESEARCH REACTOR FUEL DEVELOPMENT

Research and test reactors are used for the following activities: radioisotopes production. materials testing, training of reactor operators. neutronic studies. neutron radiography and activation analysis. In accordance with the Research Reactor Database (RRDB) of December 1994 published by IAEA [5] there were 296 research reactors in operation. 12 under construction, 8 planned and 272 shut-down in the world. In the post-war period there was a significant development on research reactors and their fuel elements. The power of these reactors varies from about 1 watt to more than 100 MW. The various types of research reactors can be grouped according to some criteria. for example, the moderators and coolants used in them (graphite moderated. light-water-moderated heterogeneous. heavy-water-moderated), or the fuel type. The fuel elements that have been used in research reactors include: uranium metal. uranium alloys, dispersions in aluminum (UAlx. U_3O_8, U_3Si_2), dispersions in graphite and stainless steel (UO_2), UZr-hydride. aqueous solutions (uranyl sulfate. nitrate or phosphate) [6]. In spite of the fact that there are a significant percentage of experimental fuel and reactor concepts. the majority is constituted by MTR and Triga reactors. The start-up of the Material Test Reactor (MTR) in Idaho in 1952. a high-flux 45 MW(th) power instalation to test materials in intense radiation field. and a plate-type dispersion-fuel reactor, gave origin to a wide group of similar research reactors employing the same fuel concept.

In the 1950s and 1960s, lower power research reactors were built around the world using MTR-type fuel elements containing low-enriched uranium fuel - LEU (< 20 mass % of ^{235}U) [7]. Although those low-power research reactors initially used low-enriched uranium fuel, the higher power research and test reactors utilized highly-enriched uranium - HEU (90-93 mass % of ^{235}U). The use of HEU has some benefits such as: it avoids the need of higher uranium concentration in the fuel, longer core resident time. higher specific reactivity and lower fuel costs. With such advantages, and since HEU was readily available. even those lower power reactors gradually were converted from LEU to HEU [8].

In 1977, however, proliferation concerns raised and determined the establishment of a new policy. That new policy established by the Carter's government conduced to severe restrictions and control of the export of HEU. Since enrichment below 20 % is considered an adequate barrier to weapons utilization, it was decided to minimize the international trade of highly enriched uranium. As a consequence of this policy, the U.S. Government established a program to make possible the fuel enrichment reduction. The U.S. Reduced Enrichment Research and Test Reactor (RERTR) Program was established by the U.S. Department of Energy in 1978 with the decision to develop proliferation-resistant fuels. The main goal of this program has been to provide means, such as the development of new fuels and design modifications, to reduce the enrichment of the fuels used for test and research reactors.

The efforts of the RERTR program are concentrated on plate-type fuels because the consumption of HEU for plate-type research reactors is much more important than for rod-type reactors. Conversion of research reactors to low enriched fuel raises some technical problems. The reduction of 93 % ^{235}U to 20 % ^{235}U requires an increase by 10 to 15 % of the ^{235}U amount in order to overcome the additional neutron absorption of the increased ^{238}U content in LEU fuel and the effects of a harder neutron spectrum. The additional uranium content (^{235}U and ^{238}U) can be accommodated by increasing the uranium density of the fuel and/or by redesigning the fuel element to increase the volume fraction in the core [9].

Since the beginning of the RERTR program, it was decided to compensate the reduction of the enrichment by increasing the uranium loading in the fuel plates. The uranium compound firstly used was the U-Al alloy. It was decided to replace this compound by another which could contain more uranium. Several uranium compounds were experimented. The development history of aluminum-based dispersion fuel could be divided into the Al-U alloy fuel period (1950s), the U_3O_8-Al and UAlx-Al dispersions period (1960s) and the high density dispersion fuel period (1980s), when UAlx-Al at 2.3 g U/cm^3, U_3O_8-Al at 3.2 g U/cm^3 and silicide aluminum dispersed fuels were developed. Silicide fuels were chosen for their higher uranium content and acceptable behavior under irradiation. Tests carried out with miniplates by the Argonne National Laboratory under the auspices of the RERTR program confirmed the good irradiation performance of the U_3Si_2 fuel. In 1988, the U. S. Nuclear Regulatory Commission approved fuels up to 4.8 g U/cm^3, allowing the conversion of the vast majority of research reactors from the use of high to low enriched uranium. The silicide fabrication experience and technology are very well described in some previous reports [10,11,12].

CERCA - Compagnie pour l'Étude et la Realisation de Combustibles Atomiques - reported recently the successful fabrication of silicide fuels with 6 g U/cm^3 employing optimized fabrication procedures. Combining thicker meat and higher volume fraction of U_3Si_2 it could be possible to reach 6.5 gU/cm^3 [13]. Nevertheless, limitations in manufacturing do not allow higher loading. Assuming that the fabrication of this fuel has acceptable yields and it is commercially viable, 6 g U/cm^3 would be today the upper limit of uranium loading for fuel plates. Besides, CERCA reported some developments using uranium nitride dispersion fuel (UN-Al) and, in spite of the higher thermal neutron absorption cross section of nitrogen, which reduces the advantages of nitrides related to silicides, the uranium densities can reach about 7 g/cm^3 for this compound [14].

However, there are several high performance reactors that require uranium loading of up to 9 g/cm^3. The new phase of the RERTR program is being dedicated to develop such very high uranium loading fuels, with special attention to very high density γ-stabilized uranium alloys.

III. DISPERSION FUEL CONCEPT

Research reactor fuel elements can be found in a variety of shapes, sizes, and compounds. The fuel enrichment have included natural uranium, under 20 % enriched uranium and 20 to 93 % enriched uranium. Any reactor that uses fully enriched uranium (93 %) can be modified to operate at 20 % fuel. Of course, the fuel enrichment will influence the design of the fuel elements and there are many compromises between operational features and economic considerations in selecting the fuel for each reactor. For example, natural uranium reactors require large amounts of fuel and use expensive heavy water or high purity graphite as moderators. Enriched fuels in the form of solid fuel elements can be used in some reactor concepts as, for example, pool or tank type research reactors. These reactors provide extreme operational flexibility, adequate neutron fluxes for most research activities, safety and economy.

Fuel elements can be assemblies of plates or rods. Nevertheless, the present review is concentrated on the plate-type aluminum-based dispersion fuels. Aluminum is used as the major construction material for a large number of research reactor fuels. The plate-type fuel consists of a fuel core in an aluminum alloy cladding. This plate-type fuel is used for most research and test reactors. The fuel core can be constituted by U-Al compounds precipitated in an aluminum matrix (cast alloy cores, concept used for the early dispersion fuels) or a uranium compound dispersed in aluminum. For special applications, other metallic or ceramic matrix could be used. The main concept involved in a dispersion fuel is to isolate the fuel particles in order to assure that an important part of the matrix remains undamaged by fission products [15]. The uranium compound particles are distributed in a thin and uniform layer inside the fuel plate, bonded to the matrix aluminum particles. The aluminum cladding, diffusion bonded to the frame and meat, assures that good heat transfer conditions are obtained and avoids the oxidation of the uranium compounds and the release of fission products to the coolant. Assuring that the fission products are contained and that good heat transfer properties are maintained, it is possible to reach very high burnups, which would be impossible in bulk fuels (UO_2 fuel pellets, for instance).

IV. FUTURE DEVELOPMENTS

The U_3Si_2 fuel compound has excellent performance under irradiation, but limited uranium density. As it seems to be impossible to increase the fuel volume fraction beyond 50-55 % with the present technology, because considerable optimization has already been performed for this methodology, the fuel plate uranium loading must be increased by changing U_3Si_2 for another higher-density fuel. The new fuel must have similar properties such as manufacturing capacity, compatibility with aluminum and stable irradiation behaviour. There are some constraints such as limited plant and process modifications in the current comercial facilities.

Many uranium compounds have density advantages over U_3Si_2: UN (14.3*/13.5**), U_3Si (15.4*/14.8**), U_6Ni (17.6*/16.9**), U_6Fe (17.7*/17.0**), U_6Mn (17.8*/17.0**). * compound specific mass in g/cm^3; **compound U-density in gU/cm^3. Nevertheless, some intermetallic compounds obtained from peritetic /peritectoid transformations have shown poor irradiation behaviour [16]. Uranium nitride has an excellent irradiation performance but a small density advantage over U_3Si_2.

Alloys with stable uranium gamma phase have better performance than uranium alpha phase or than peritectic compounds susceptable to amorphization. Modifications of uranium metal to achieve higher irradiation stability involve stabilization of gamma phase at low temperatures. There are some metastable gamma phase stabilizers that present low neutron capture cross section: Zr, Nb, Ti and Mo. These alloys have shown good irradiation performance under fast reactor conditions [4], under high temperature and moderate burnup. There are not data available for fuel performance under conditions that are found in thermal research reactors (low temperature and high burnup). On the one hand, it must be considered that the γ phase stabilizing requires high alloys. On the other hand, high alloys mean lower U density. The most promising single alloys are from the U-Mo group, thanks to its relatively large range of gamma phase in addition to the relatively low neutron capture cross-section of the molybdenum

Besides the irradiation behaviour the problem of the fuel-matrix interaction must be considered. In case of excessive reaction between the potential alloys and the aluminum matrix, other materials must be investigated (magnesium matrix/aluminum cladding or zirconium both matrix and cladding) by the renewed RERTR program [4]. The first test will focus on some physical matallurgy experiments related to fuel particle-matrix interactions to establish the best candidate alloy series.

The alloys must be powderized, homogeneized with aluminum, compacted and roll bonded in order to preserve the present technology and minimize the impact in the fabrication costs. To retain the cubic γ U phase in a metastable state it is important a rapid solidification method. If the metastable γ phase can be maintained during fuel element fabrication and irradiation for U-Mo alloys, and if these alloys have good thermal compatibility with aluminum, they would be a prime candidate as a dispersion fuel for research reactors. A centrifugal atomization process has been applied in the U-Mo powder production. The atomized powder has spherical shape, narrow size distributions, average diameter of 83 μm, high density and fine grain structure with isotropic γ - U phase. Fuel rods constituted by U-10wt.% Mo have shown good thermal compatibility with Al matrix and mantain a microstructure stability, as reported in [17].

V. CONCLUSIONS

Although very high U density fuels are not necessary for the program of converting IEA-R1 Brazilian Research Reactor from HEU to LEU [18] and the planned upgrade and power increase from 2 to 5 MW, it would be very important to establish close cooperation with the Institutions involved and to participate in the new development cycle.

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