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PREPARATION OF FIRST MARK IA FUEL ALLOY FOR EBR-II

by

W. E. Miller, G. J. Bernstein,
J. F. Lenc, and R. C. Paul

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Chemical Engineering Division

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PREPARATION OF FIRST MARK IA FUEL ALLOY FOR EBR-II

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I. INTRODUCTION

This report describes the preparation of the Mark IA fuel alloy and serves as a guide for preparation of EBR-II fuel alloys.

The fuel pins for the first core (FC) loading of the EBR-II reactor were made from 859 kg of uranium-5 w/o fission* alloy in 1959.¹ The uranium enrichment in the fuel was $48.1 \pm 1/2$ w/o ^{235}U . After the initial period of reactor operation, it became desirable to use fuel with a higher uranium enrichment. Therefore, in late 1965, 200 kg of uranium-5 w/o fission fuel containing $52.2 \pm 1/2$ w/o ^{235}U in the uranium portion of the fuel

was prepared for the reactor. This latter fuel alloy was designated Mark IA fuel to differentiate it from FC fuel. Table I shows the nominal compositions of the two fuel alloys for the FC and Mark IA fuels.

TABLE I. Nominal Composition
of EBR-II Fuel Alloys

Element	Fuel-alloy Composition (w/o)	
	FC	Mark IA
Uranium, total		
^{235}U	48.13 ^a	52.18 ^a
^{234}U , ^{236}U , ^{238}U	51.87 ^a	47.82 ^a
Molybdenum	2.46	2.46
Ruthenium	1.96	1.96
Rhodium	0.28	0.28
Palladium	0.19	0.19
Zirconium	0.10	0.10
Niobium	0.01	0.01
Total	100.00	100.00

^aWeight percent of isotopes based upon total uranium in fuel alloy.

The equipment and procedures used to prepare the Mark IA fuel alloy were essentially the same as those employed in preparing the FC fuel alloy.¹ Some differences did arise, however, as a result of the larger batch sizes used in preparing the Mark IA fuel. The average weight of a charge to the melt crucible for preparing the FC fuel alloy was 10.4 kg, whereas the average charge weight for preparing the Mark IA

fuel alloy was 13.8 kg. The larger melts of the Mark IA fuel alloy were cast into double-compartment ingot molds rather than into the single-compartment molds used in preparation of the FC fuel alloy.

*Fission is a general name given to a variable mixture of fission-product elements (atomic numbers 40 thru 46) which, when alloyed with uranium, impart to the alloy desirable metallurgical properties and radiation stability.

The size of the ingots cast was limited by the capacity of the fuel-pin injection-casting furnace. Therefore, casting double ingots reduced the number of runs required to prepare fuel alloy by a factor of two, and the ingots were small enough to fit the casting furnace. The crucibles used for the preparation of the alloy were lime-stabilized, pressed and sintered zirconia crucibles having an inside diameter of $5\frac{1}{8}$ in. and an inside depth of $8\frac{1}{2}$ in. The weight of a typical crucible was about 9.6 kg.

II. PREPARATION OF FUEL ALLOY

Prior to preparation of the Mark IA alloy, the equipment and operating procedures to be used were reviewed and approved by Argonne National Laboratory's Criticality Hazards Control Committee. The Committee noted that strict adherence to the proposed sequence of operations assured the presence of only one 13.8-kg batch of Mark IA fuel alloy in the fuel-alloy preparation area at any given time. This quantity allowed for a safety factor of 2.3 based on a 32-kg batch of 50 a/o ^{235}U enriched fuel alloy that is fully water-reflected.

The sequence of the steps involved in conducting a fuel-alloy preparation run are shown schematically in Fig. 1. These steps are described below with the numerals in parentheses identifying the specific steps in Fig. 1.

Steps ①, ②, and ③ completed before 10:00 a.m. each day.

Fig. 1. Numerical Sequence of Steps Involved in Preparation Runs of Mark IA Fuel Alloy

At the beginning of each day, the ingot that had been cast the preceding day, and the melt crucible and skull, were weighed, bagged, and removed from the building by Special Materials and Services (SPM) personnel (1). The uranium was then delivered for the current day's run (2) and, after a weight check, was added to a new crucible along with the noble-metal charge that had been prepared the previous day and stored in the building (3). The noble-metal charge was based upon information received the preceding day (6) as to the amount of uranium that would be delivered for the current day's run. The charge was then melted (4), cast into a mold, and allowed to cool overnight. Information (6) was received each day from SPM personnel on the weight of uranium to be delivered and the amount of alloy removed from the building the previous day (5). This information (6) was recorded and checked against weighings made in the building before charging each run so that a continuous material balance could be maintained (7).

The fuel alloy was prepared in a furnace that had been installed within a three-module glovebox used in an earlier experimental program. Since the preparation of the fuel alloy did not require an inert atmosphere around the outside of the furnace, one glovebox window was removed to provide direct access to the furnace.

The furnace was a prototype of the units currently in service in the EBR-II Fuel Cycle Facility in Idaho.^{2,3} Figure 2 shows the furnace in the glovebox with its bell-jar cover removed. During furnace operation, the bell jar was secured to the furnace with a fusible-metal seal.⁴

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Fig. 2. Work Area Used for Preparation of Mark IA Fuel Alloy

The first step in conducting a fuel-alloy preparation run was to melt the fusible seal and open the furnace by raising its bell-jar cover out of the seal. (The ingot that had been poured on the previous day remained in the furnace overnight for cool-down with the seal heater switch locked in the "off" position to provide secure overnight storage for the enriched fuel alloy.) After the furnace was opened, the melt crucible containing the skull of unpoured metal and oxide, and the graphite mold containing the fuel alloy ingot from the previous day's run, were removed from the furnace.

Suitable respirators and rubber gloves were worn by the operators during the handling of the melt crucible and the mold to prevent radiation exposure by ingestion or by contact with the skin. Personnel of the Industrial Hygiene and Safety Division were present during all operations to monitor the operators and the work area. The melt crucible and skull were weighed, sealed in a plastic bag, and removed from the work area by a member of the Special Materials and Services Division.

108-9018

Fig. 3. Double-compartment Mold Used for Preparation of Mark IA Fuel-alloy Ingots

The CS grade* graphite double-compartment mold (shown in Fig. 3) containing the fuel-alloy ingot was removed from the furnace and placed upside down on a flat stainless steel plate. The ingot-bearing mold was then lifted manually about 6 in. above the plate and forcibly struck against the plate. Generally, one blow was sufficient to release the ingot from the mold.

Each half of the ingot was a tapered cylinder, $3\frac{1}{2}$ in. in diameter at the top, $3\frac{1}{8}$ in. in diameter at the bottom, and having a 3° wall taper. The height of the ingot was about $2\frac{1}{4}$ in. Two types of protrusion samples were formed as integral parts of each half of an ingot in the casting process. Suitable cavities were machined in the bottom of each compartment of the double-compartment mold for this purpose. One such cavity is visible in the bottom of one of the compartments of the mold shown in Fig. 3. One type of sample was in the form of a small tapered cylinder weighing about 2.5 g. This type of sample was used for spectrophotometric and

*National Carbon Company designation.

isotopic mass-spectrographic analyses. The other type was in the form of a flat, trapezoidal-shaped blade of metal with the two larger surfaces each about 1 in. square. This type of sample, which weighed about 20 g, was used for X-ray spectrometric analyses.

Figure 4 is a bottom view of a typical Mark IA fuel ingot with its integrally cast blade samples still intact. The small tapered protrusion samples have been removed from this ingot. The protrusion samples were broken off the ingot by a blow with a hammer and cold chisel. This operation was performed within the small portable glovebox shown in Fig. 5. The small glovebox was used to contain the samples, which flew in random directions when they were broken from the ingot. The small glovebox was placed over an unattached stainless steel plate. A mold, ingot, and samples were placed into or removed from the box by merely tilting the box as shown in Fig. 6. After removal of the samples, the ingot and samples were weighed, sealed in plastic bags or in sample bottles, and then removed from the fuel-alloy preparation area by a member of the SPM Division.

After the ingot had been removed from the area, SPM personnel broke the ingot into two approximately equal halves by fracturing the web section in a hydraulic press. Each half-ingot was used as a charge to an injection casting furnace crucible. In the casting furnace (operated by the Metallurgy Division), each half of the ingot was remelted, and Mark IA fuel pins were injection-cast from the melt.⁵

After removal of the ingot, samples, and skull-bearing crucible from the fuel-alloy preparation area, a member of the SPM division brought the uranium charge for the next fuel-alloy preparation run into the area. The

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Fig. 4. Bottom View of Typical Mark IA Fuel-alloy Ingot

Fig. 5.

Work Area for Removal of Protrusion
Sample from Mark IA Fuel-alloy
Ingot

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Fig. 6. Recovery of Ingot Protrusion Sample

noble-metal elements that had been weighed on the previous day were then removed from storage. Both the uranium and the alloying elements were weighed individually to the nearest 0.1 g before they were charged to the

crucible. The total charge was then weighed as a check. Table II lists the weights of the various elements used in a typical charge.

TABLE II. Typical Charge for Preparation
Run of Mark IA Fuel Alloy

Element	Weight (g)
Uranium, enriched (93.15 w/o ^{235}U)	5,119.7
Uranium, diluent (25.57 w/o ^{235}U)	7,858.5
Molybdenum	349.7
Ruthenium	267.8
Rhodium	38.3
Palladium	26.0
Zirconium	16.1
Niobium	1.4
Total	13,677.5

The procedure used in determining the charge weights of the various elements in the FC fuel-alloy preparation runs¹ was also used in the preparation of the Mark IA fuel. Molybdenum and zirconium were added to the charge in amounts slightly in

excess of those required to obtain the nominal fuel alloy composition shown in Table I. As a result, the total uranium content of the charge was 94.88 w/o, instead of the nominal 95.0 w/o for the fuel alloy. The slight excess of molybdenum and zirconium was added because these two elements did not alloy quantitatively with the uranium in previous uranium-fissium alloy preparation runs. (In the previous runs, the molybdenum and zirconium contents of the unpoured metal skull that remained in the melt crucible were greater than their contents in the poured ingot.)

The noble metals (molybdenum, ruthenium, rhodium, and palladium in the form of pressed pellets) were loaded into the crucible first. Zirconium rod and niobium powder were then added to the noble-metal elements in the bottom of the crucible. The uranium charge was in the form of broken biscuit material and was made up of metal of two different enrichments, 93.15 and 25.57 w/o ^{235}U . The required amount of uranium metal of each enrichment was loaded into the crucible on top of the alloying elements.

After the crucible was loaded and weighed, it was inserted into the furnace susceptor by hand. Since the loaded crucible weighed about 23 kg, sliding it into an inclined susceptor was safer than lowering it into the susceptor in a vertical position. The latter procedure would have meant some risk of dropping the crucible and cracking both the crucible and the furnace susceptor. Therefore, the furnace assembly was tilted as shown in Fig. 7 to facilitate insertion of the loaded crucible. The furnace was tilted with a jack located under the base plate. The jack was connected through a bellows seal to a rack-and-pinion drive assembly on the furnace support frame. Next, a Fiberfrax* furnace-top insulating cover was placed in a holder on the furnace frame, and the double-compartment mold was positioned as shown in Fig. 7, so that the molten fuel alloy would pour into the center of the compartment closest to the furnace assembly. The molten

*A ceramic fiber made of alumina and silica by the Carborundum Company.

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Fig. 7. Furnace Assembly Used for Preparation of Mark IA Fuel Alloy

charge also was obtained from SPM personnel, and the noble-metal elements for the next day's charge were weighed on the basis of this information. This work was conducted concurrently with melting and casting the fuel alloy for the current day's run.

The Mark IA fuel alloy was melted and subsequently cast in the following manner: First the furnace was evacuated to less than 1 Torr. Then the valve in the vacuum line connecting the furnace to the vacuum pump was closed to isolate the furnace from the vacuum pump. If no substantial increase (<0.1 mm Hg) in the furnace pressure was detected after 5 min, the furnace was considered to be sufficiently vacuum-tight for the fuel-alloy melting operation.

Argon was introduced into the furnace until the furnace pressure was slightly above atmospheric, and the furnace was then inductively heated to a crucible temperature of 1400°C in about $2\frac{1}{2}$ hr. The crucible temperature was measured by means of a top reference thermocouple positioned in the center of the Fiberfrax insulating cover (see Fig. 7), and a bottom reference

alloy could flow from this compartment into the other compartment through a mold gate. The furnace assembly with the loaded crucible installed was then tilted back to its upright position so that the crucible was vertical in the furnace.

Finally, the bell-jar cover was lowered into the molten metal seal on the furnace base plate,⁴ and a vacuum-tight seal was made between the bell jar and the base plate by allowing the molten-metal seal to freeze.

The weights of the ingot and samples which were determined when the materials were removed from the furnace were compared with the weights subsequently obtained by SPM personnel in another area. Agreement in these weights provided a check on the material balance for the run. The exact weight of the next day's uranium

thermocouple located in the bottom of the furnace assembly. These reference thermocouples had been calibrated in advance so that actual melt temperatures could be determined within $\pm 10^\circ\text{C}$.

The crucible was maintained at 1400°C for 2 hr to melt the charge. The molten charge was poured into the double-compartment graphite mold by smoothly tilting the furnace assembly to a horizontal position. A thermocouple attached to the graphite mold was used to verify the presence of the molten charge in the mold. As soon as the molten charge was poured out of the melt crucible, the furnace assembly was tilted back to its original vertical position.

III. RESULTS OF ALLOY PREPARATION RUNS

A. Pouring Yields

After the molten charge was poured in a Mark IA fuel-alloy preparation run, a skull of unpoured metal remained in the zirconia melt crucible. The weight of the ingot cast in a given run, therefore, was less than the weight of metal charged to the crucible. The pouring yield was determined for each of the 15 Mark IA fuel-alloy preparation runs by the following equation:

$$\text{Pouring yield (percent)} = \frac{\text{Weight of ingot}}{\text{Weight of metal charge}} \times 100.$$

Table III presents the average pouring yield and average ingot weight for the 15 runs.

TABLE III. Yield Data for Preparation Runs of Mark IA Fuel Alloy

Number of Runs	Average Pouring Yield (%)	Maximum Yield (%)	Minimum Yield (%)	Average Ingot Weight (kg)
15	96.2 ± 0.6^a	97.8	95.3	13.273

^aAll values of uncertainty used in this report are based on the equation

$$\sigma = \sqrt{\frac{\sum d^2}{n-1}},$$

where σ is the degree of uncertainty and d is the deviation of the individual determination from the arithmetical average of n determinations.

The average pouring yield of 96.2% for the Mark IA runs compares favorably with the average pouring yield of 96.8% reported for 85 FC fuel-alloy preparation runs.¹ However, since the skull weight is roughly proportional to the ratio of the crucible surface area to the charge weight for a given run, it was anticipated that the lower ratio of surface area to charge weight in the Mark IA fuel-alloy preparation runs would result in higher pouring yields than those achieved in the FC fuel-alloy preparation runs. There are two possible explanations for the lower pouring yields obtained in the Mark IA fuel-alloy preparation runs. Experience in the preparation of the FC fuel alloy indicated that the pouring yield tended to increase as the number of runs increased. This increase in yield was attributed to a decrease of gaseous impurities in the furnace components as the runs progressed. The degassing was a result of repetitive use of a furnace over a long period of time. Since only 15 runs were made in the preparation of the Mark IA fuel alloy, as compared with 85 runs for the FC fuel alloy, this degassing effect was not as pronounced in the Mark IA runs as it was in the FC runs.

Another possible explanation for the difference in yields is that the Mark IA and FC fuel alloys were prepared from different batches of enriched and diluent uranium, which may have contained different levels of oxygen impurity. No information is available on the oxygen content of the uranium used in the preparation of the two fuel alloys.

B. Isotopic and Total Uranium Content

Specifications for the Mark IA fuel alloy required an enrichment of 52.18 ± 1.2 w/o (52.5 ± 1.2 a/o) ^{235}U in the uranium present in the alloy. In addition, a total uranium content of 95.0 ± 1.0 w/o was specified for the alloy.

The average isotopic uranium content of the uranium in the Mark IA fuel alloy prepared in 15 runs was 0.519 ± 0.009 w/o ^{234}U and ^{236}U combined, 47.30 ± 0.07 w/o ^{238}U , and 52.18 ± 0.07 w/o ^{235}U . The total uranium concentration in the alloy averaged 94.7 ± 0.6 w/o. These values are averages of mass-spectrographic analyses of ingot protrusion samples from each of the 15 runs. Seventeen analyses were performed in all, because both halves of two of the ingots were sampled to determine the isotopic homogeneity between the two halves of double ingots. This method of sampling was used because there was some concern over possible segregation of the alloying elements between halves of an ingot. Table IV presents the analyses of the ingots from these two runs. All the ingots met the specifications set up for Mark IA Fuel.

TABLE IV. Isotopic Uranium Analyses for Both Halves of Ingots from Two Preparation Runs of Mark IA Fuel Alloy

Run No.	Ingot Half No. ^a	Uranium Content (w/o) ^b		
		²³⁴ U and ²³⁶ U	²³⁸ U	²³⁵ U
6	1	0.534	47.34	52.12
6	2	0.532	47.32	52.09
14	1	0.513	47.26	52.22
14	2	0.513	47.28	52.21

^aIngot half No. 1 solidified in the mold compartment into which the melt was poured directly. Ingot half No. 2 solidified in the other mold compartment into which the melt flowed from the first mold compartment through the mold gate.

^bBased on mass-spectrographic analyses of ingot protrusion samples.

C. Noble-metal Content

The noble-metal content of the 15 Mark IA fuel-alloy ingots was determined by spectrophotometric analyses of one of the two protrusion samples from each half of the ingots. Additional analyses for molybdenum and ruthenium were obtained by X-ray spectrometric analyses of the other protrusion sample from each half-ingot. Table V presents the averaged molybdenum and ruthenium contents for ingot halves No. 1 and 2 of all 15 ingots. The results indicate that there was no segregation of molybdenum and ruthenium between ingot halves for any of the runs.

TABLE V. Averaged Molybdenum and Ruthenium Content of Mark IA Fuel Alloy Based on X-ray Spectrometric Analyses of Ingot Protrusion Samples from 15 Runs

Ingot Half No.	Element Content (w/o)	
	Molybdenum	Ruthenium
1	2.48 ± 0.09	2.01 ± 0.03
2	2.46 ± 0.06	2.01 ± 0.02

Table VI lists the fissium contents of one ingot half from each of two alloy preparation runs. The table also includes the molybdenum and ruthenium X-ray spectrometric analyses for the two half-ingots.

The data presented in Table VI indicate that the palladium and zirconium contents of the two half-ingots were essentially as specified, i.e., 0.19 and 0.10 w/o, respectively. The results of the spectrophotometric analyses

presented in the table were about 5% higher for molybdenum and about 5% lower for ruthenium than the results of X-ray spectrometric analyses. The differences between the results of spectrophotometric and X-ray spectrometric analyses for molybdenum and ruthenium appear to be characteristic of the different analytical methods used, since these differences are greater than the deviations resulting from X-ray spectrometric analysis alone (see Table V). Further evidence of these differences is described below.

TABLE VI. Typical Fissium Content Based on Analyses of One Half-ingot from each of Two Preparation Runs of Mark IA Fuel Alloy

	Fissium Content (w/o) ^a	
	Run 9; Ingot Half No. 1	Run 12; Ingot Half No. 1
Mo	2.61 2.47 ^b	2.63 2.55 ^b
Ru	1.92 2.00 ^b	1.84 2.02 ^b
Rh	c	c
Pd	0.19	0.18
Zr	0.11	0.10
Nb	c	c

^aBased on spectrophotometric analyses of ingot protrusion samples, unless otherwise indicated.

^bBased on X-ray spectrometric analyses of ingot protrusion samples. Results of duplicate analyses are shown.

^cNo analyses were made for rhodium and niobium; however, other investigations have shown that these two elements alloy almost quantitatively.^{1,7}

Fuel pins that were injection-cast from five Mark IA fuel-alloy ingots⁶ were checked by spectrophotometric analysis for molybdenum, ruthenium, rhodium, and palladium. Table VII shows the averaged results of the analyses for pins from each half of the five fuel-alloy ingots. The averaged results in this table indicate there was no segregation of the rhodium and palladium alloying elements between pins cast from two half-ingots in each run.

TABLE VII. Averaged Content of Certain Fissium Elements in Fuel Pins Injection-cast from Each Half of Five Mark IA Fuel-alloy Ingots

Ingot Half No.	Element Content (w/o) ^a			
	Molybdenum	Ruthenium	Rhodium	Palladium
1	2.59 ± 0.05	1.93 ± 0.07	0.27 ± 0.02	0.20 ± 0.00
2	2.65 ± 0.08	1.91 ± 0.10	0.27 ± 0.02	0.20 ± 0.00

^aBased on spectrophotometric analyses of fuel-pin samples.

Table VIII compares the averaged molybdenum and ruthenium analyses of the five Mark IA fuel-alloy ingots with the averaged molybdenum and ruthenium analyses of injection-cast fuel pins from the same five ingots. The values for the ingots in this table were based upon X-ray spectrometric analyses, whereas the values for the fuel pins were based upon spectrophotometric analyses. The deviation between the X-ray spectrometric analyses and the spectrophotometric analyses in Table VIII is similar to the deviation reported in Table VI, although the X-ray spectrometric analyses in Table VI were based on ingot protrusion samples and the spectrophotometric analyses in Table VIII were based on fuel-pin samples. In spite of the difference in the source of the samples, the data in Table VIII appear to confirm that the two methods of analyses do give different results for the same molybdenum and ruthenium contents.

TABLE VIII. Comparison between Averaged Molybdenum and Ruthenium Analyses of Halves of Five Mark IA Fuel-alloy Ingots and Fuel Pins Injection-cast from Halves of the Five Ingots

	Element Content (w/o)			
	Ingot Half No. 1		Ingot Half No. 2	
	Ingot Protrusion Sample ^a	Fuel-pin Sample ^b	Ingot Protrusion Sample ^a	Fuel-pin Sample ^b
Mo	2.42 ± 0.09	2.59 ± 0.05	2.45 ± 0.03	2.65 ± 0.08
Ru	2.02 ± 0.04	1.95 ± 0.07	2.02 ± 0.02	1.91 ± 0.10

^aBased on X-ray spectrometric analyses.

^bBased on spectrophotometric analyses.

IV. CONCLUSIONS

1. Fifteen uranium (52.2 ± 1.2 w/o enriched)-fissium Mark IA fuel-alloy ingots were prepared. All the ingots met the composition specifications set up for Mark IA fuel. Fuel pins were injection-cast from these ingots for use in the core of the EBR-II reactor.
2. The feasibility of successfully casting two homogeneous half-ingots in one fuel-alloy preparation run was demonstrated.
3. Spectrophotometric analyses showed 5% higher molybdenum content and 5% lower ruthenium content than X-ray spectrometric analyses of the same Mark IA fuel-alloy ingots.

V. PREPARATION OF SUPPLEMENTARY EBR-II FUEL ALLOY

In addition to the 200 kg of Mark IA fuel alloy prepared in the runs described above, 27 kg of uranium-fissium alloy with a higher ^{235}U

enrichment (69.8 ± 1.2 w/o) was prepared. The latter fuel alloy (designed Special Enriched Fuel Alloy) was made in two preparation runs that were similar to the 15 Mark IA fuel-alloy preparation runs. The Special Enriched Fuel Alloy was prepared to make EBR-II fuel pins which could achieve a desired burnup in a shorter period of time than the Mark IA fuel-alloy pins. Examination of the Special Enriched Fuel Alloy pins after the desired burnup had been achieved would permit valid predictions of the condition of Mark IA fuel-alloy pins after the same burnup. This information was required before the Mark IA fuel-alloy pins were allowed to reach the desired burnup.

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