MORNING LIGHT CLEANUP AND RECOVERY OPERATION: SIMULATION STUDIES OF POSSIBLE REACTOR FUELS

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ABSTRACT

The nuclear fuel for Cosmos 954, the orbiting Russian reactor that broke up on reentry during January of 1978, has been identified as a U-Mo alloy containing about 10 wt% molybdenum. Identification was based on a combination of simulation studies at LLL, examination of fuel debris at Whiteshell Nuclear Research Establishment (WNRE), Pinawa, Manitoba, and reactor technology knowledge. In the LLL simulation studies, mixtures of uranium, molybdenum, and UO_2 were heated under conditions that simulated reentry and then examined by scanning electron microscopy, energy dispersive spectrometry, and x-ray diffraction. These studies indicated metallic behavior and suggested a U-Mo alloy.

The identification was useful in assisting the Canadians in recovery, cleanup, and health/safety activities associated with the radioactive debris, which was scattered over a wide region of the Great Slave Lake.

INTRODUCTION

When Cosmos 954, the orbiting Russian reactor, broke up on reentry, it scattered radioactive debris over a wide region of Great Slave Lake, Canada. Lawrence Livermore Laboratory was asked to participate in the cooperative American and Canadian cleanup operation known as Morning Light. The Canadians wanted to know the composition of the fuel so that they could reconstruct the reactor design and subsequently predict amounts and dispersion of the radioactive debris.

Our initial opinions on the identity of the fuel were based on preliminary analysis of radioactive microspheres picked up in contaminated areas immediately following the reentry. The Whiteshell Nuclear Research Establishment (WNRE), which was responsible for analysis of the debris, detected UO_2 and molybdenum in the debris. It was first thought that the fuel might be a UO_2- or UC-Mo cermet because a molybdenum cermet would improve thermal conductivity and reduce swelling from radiation damage. Another alternative was a U-Mo alloy. Uranium-molybdenum alloys were logical candidates because of their many favorable properties (easy fabrication, reasonable tensile properties, corrosion resistance, and a stabilized gamma phase). As more information became available, we narrowed the choices to two fuel systems. UO_T-Mo and U-Mo.

Knowledge of the fuel was a key piece of information because it would tell much about the design of the reactor. Cermet fuel elements (UO_2-Mo system) generally have a significantly lower thermal conductivity than alloy elements (U-Mo system). Fuel systems of lower thermal conductivity (UO_2-Mo, UN-Mo, etc.) suggest a Topaz design. [1,2] Higher conductivity systems (U-Mo, UC-Nb, etc.) suggest a Romashka design. [3,4] Another fuel system, Mo-UN, could also be used in the Topaz design, but the Russians have not shown any significant interest in the nitride system. In contrast, they have been active in developing the alloy (U-Mo), oxide (UO_T-Mo), and carbide (U_xC_y-Mo) systems. [5,6]

After several trips to WNRE for technical discussions, U-Mo and UO_T-Mo were still considered the most likely fuel systems. Further analysis of debris and/or simulation studies were needed to choose between the two possibilities. At that time we suggested further evaluation of the fuel debris at WNRE and simulation studies at LLL. The aim of the simulation work was to subject likely fuel compositions to the heat and pressure conditions experienced by the actual fuel during breakup and reentry and compare the resulting microstructures with the microstructures present in the recovered microspheres. For example, if dendritic phases such

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as those found in the fuel debris were duplicated in our simulation studies, this would be a good clue to the identity of the fuel.

Secondary objectives of these studies were to estimate the approximate composition and time sequences and conditions of breakup and reentry.

SIMULATION STUDY PROCEDURES

Sample Preparation

We prepared specimens from high-purity molybdenum, uranium, and UO_2 (depleted uranium in every case) for heat treatments at various temperatures and in various environments (see Table 1). Two preparation techniques were selected to simulate reentry conditions.

The first technique was vacuum (10 mPa) heat treatment (VHT) to simulate the low pressure and low thermal conductivity of the reentry environment. Molybdenum foil (11 mm thick) was formed into crucibles to hold uranium chips or UO_2 microspheres and then vacuum heat treated inside covered BeO crucibles. The foil was both the source of molybdenum for alloying as well as the reactant

Table 1. Summary table of heat treating conditions and x-ray diffraction results for UO_2-Mo and U-Mo specimens.

Specimen	Composition, wt%			Heat treatment			X-ray diffraction detected phases[a]			Surface condition	DPH (200 g load)
	Mo	U	UO$_2$	Temp, °C	Time, min	Environment	UO$_2$	Mo	Others		
UO$_2$ microsphere			bal.	-	-	-	S	ND	ND	As-received	-
1	8		bal.	2200	10	Vac	.				=
2	15		bal.	2200	10	Vac	ND	S	ND	Inside Mo boat	=
2	15		bal.	2200	10	Vac	ND	S	ND	Outside Mo boat	=
3	15		bal.	2200	40	Sealed Mo can	S	S	unk	Polished cross sections	=
4	25		bal.	2200	40	Sealed Mo can	S	S	ND	Polished cross sections	-
5	8	bal.		1300	40	Vac	S	ND	UO (M)	Top surface	250
5	8	bal.		1300	40	Vac	S	ND	UO (W)	Bottom surface	
6	15	bal.		1300	40	Vac	S	ND	UO (W)	Top surface	410-380
6	15	bal.		1300	40	Vac	S	ND	UO (T)	Bottom surface	
7	15	bal.		arc-melted		Argon	ND	W	MoU$_x$ (S)	Polished cross sections	400-270
8	25	bal.		1300	45	Vac	S	ND	UO (M) MoU$_x$ (T)	Top surface	380-375
8	25	bal.		1300	45	Vac	S	T	UO (W)	Bottom surface	
9	25	bal.		1600	5	Vac	S	W	unk	Top surface	510-370
9	25	bal.		1600	5	Vac	S	ND	MoU$_x$ (T)	Bottom surface	
10a	25	bal.		arc-melted		Argon	ND	M	MoU$_x$ (S)	Polished cross sections	400-360
10e	25	bal.		1855	2 h	Vac	W	M	MoU$_x$ (S)		670-700
11	40	bal.		1300	45	Vac	T	T	MoU$_x$ (S)	Top surface	390-360
11	40	bal.		1300	45	Vac	S	T	UO (W)	Bottom surface	
12	40	bal.		1600	5	Vac	S	M	MoU$_x$ (T)	Top surface	500-390
12	40	bal.		1600	5	Vac	S	S	MoU$_x$ (T)	Bottom surface	
13	25	bal.		arc-melted		Argon	S	S	No	Top and bottom surface	-

S – strong intensity
ND – none detected
M – medium intensity
W – weak intensity
T – trace
unk – unknown lines detected

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in the study of the rate of reaction with uranium and UO_2. Complete reaction of the molybdenum foil was required to achieve the alloy composition figures listed in Table 1. Only in the case of 8 wt% Mo did the alloying reaction go to completion.

In the second preparation technique, U-Mo and UO_2-Mo specimens were arc melted in an argon atmosphere on a chill block. Molybdenum foil was wrapped around uranium chips or UO_2 microspheres and melted with a tungsten arc electrode. Melting was repeated several times and the specimens or buttons turned over after each melting to improve mixing. This technique provided higher melting temperatures (> 2200°C), faster quenching rates on the surface in contact with the chill block, and improved mixing action between reactants during melting.

UO_2-Mo

Two initial sample compositions (8 and 15 wt% Mo) were selected as being reasonable in light of the debris compositions reported by WNRE.[7] Because of the high vapor pressure [8] of UO_2, UO_2 microspheres in molybdenum foil that were vacuum heated to 2200°C in BeO crucibles vaporized before there was any reaction. To contain the UO_2 above 1600°C, the heat treatments were repeated after sealing higher UO_2 compositions in molybdenum containers lined with BeO.

A final attempt to react UO_2 with molybdenum was made by arc melting UO_2 wrapped in molybdenum foil. The molybdenum was repeatedly melted to ensure contact of molten molybdenum with the UO_2 microspheres. These experiments were abandoned when we found there was no significant reaction between UO_2 and molybdenum under these extreme conditions (see *Results and Discussion*).

U-Mo

Alloy specimens (8, 15, 25, and 40 wt% Mo) were prepared by vacuum (10 mPa) heat treatments at 1300 or 1600°C (see Table 1). Molybdenum foil (0.11 mm thick) was formed into a rectangular boat and a piece of uranium was melted in this boat. The reaction rate of uranium with the molybdenum boat was compared at various temperatures and times. The heating rate was approximately 7°C/min and the cooling rate 100°C/min.

Two compositions, U-15 wt% Mo and U-25 wt% Mo, were arc melted in an argon atmosphere. Each button was inverted and remelted several times to improve its homogeneity. The U-25 wt% Mo button was cut into four equal pieces. Each piece was vacuum heat treated under different conditions: (1) 1600°C for 1 h, (2) 1600°C for 2 h, (3) 1600°C for 4 h, and (4) 1855°C for 2 h, respectively. Treatment (4) resulted in a 13.5% weight loss because of uranium vaporization.

Analysis

Metallography

Each heat-treated specimen was sectioned with a slow-speed, thin-slicing diamond saw. The cross sections were mounted in epoxy and polished on cloth wheels charged with diamond paste. Each cross section was examined before and after etching. Uranium-Mo specimens were electroetched in oxalic acid for ~5 s (20 V and 1 A). Uranium oxide-Mo specimens were immersion etched in lactic acid-HNO_3-HF (3:1:1 by volume). The microstructures of these cross sections were compared with the microstructures of debris provided by WNRE.

Microhardness

Diamond pyramid hardness (DPH) was measured on various regions of each of the mounted specimens. Microhardness values were calculated from the Vickers formula:

$$DPH = HV = 1854 \, P/d^2,$$

where

HV = Vickers hardness in kg/mm^2
P = Load in grams
d = Av length of the diagonals in micrometers.

Specimens were also tested with a Leitz microhardness tester under a 200 g load.

The microhardnesses (DPH, 200 g) of various regions on all specimens were measured and are summarized in Table 1. The values are comparable to published microhardness data.[9] Insufficient data were obtained on the fuel debris to make meaningful comparisons. The trends observed in Table 1 of higher microhardness with increasing molybdenum contents (above 8 wt%) and higher heat treating temperatures are consistent.

Scanning Electron Microscopy (SEM) with Energy Dispersive Spectrometry (EDS)

Heat-treated specimens were examined by SEM and EDS after metallographic examination. Features of interest were identified in the SEM and then analyzed with EDS for elemental composition. Because niobium and molybdenum, elements of interest, have EDS peaks (217 vs 230, respectively) close to each other, a series of tests was run on

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high-purity standards with uranium matrix before evaluating the heat-treated specimens. Results of calibration tests are contained in *Appendix A*.

The EDS unit was made by Ortec and had a model 6240B multi-channel analyzer and solid-state Si(Li) drifted crystal detector. Three accelerating voltages were evaluated in spot and scan mode. An area approximately $1.5 \mu m$ in diameter was analyzed in the spot mode, while the entire area in the field of a given magnification was analyzed in the scan mode. For example, in the scan mode at $100\times$ magnification, the scanned area is approximately $520 \mu m^2$ (see Ref. 10).

X-Ray Diffraction

The X-ray diffraction results in Table 1 were taken on three general types of surfaces:

1. The top surface of the melted button, which was rough and curved.
2. The bottom surface (relatively flat) of the melted button.
3. Metallographic polished cross sections of the arc-melted buttons.

For specimens vacuum heat treated, the bottom surface was in contact with a BeO crucible during vacuum heat treatment. For specimens arc-melted, this surface was in contact with a water-cooled copper chill block. The top and bottom surfaces were analyzed by x-ray diffraction techniques to identify surface layers observed during metallographic examinations. These combined results give a rough estimate of oxidation rates for various compositions at different temperatures and times in vacuum (10 mPa).

RESULTS AND DISCUSSION

Analysis at WNRE

Analysis at WNRE[11] shows that the microspherical debris from the fuel core contained uranium and molybdenum (see Fig. 1) with an outside crust of UO_2 (see Fig. 2). Uranium and molybdenum were determined by EDS while the crust was also identified by x-ray diffraction. Appropriate hot cell equipment was not available at WNRE to determine the chemical states of the uranium (carbide, oxide, nitride) and molybdenum in the central portion of the microspheres. Windowless analysis for nitrogen, carbon, and oxygen was needed to identify the core as UN-Mo, UC-Mo, or UO_2-Mo, respectively.

Most of the microspheres appeared to be similar. Dendrites were a very prominent feature of the microstructure (Fig. 3). In the dendritic regions, the light and dark areas had very different ratios of

Light regions are high in Mo

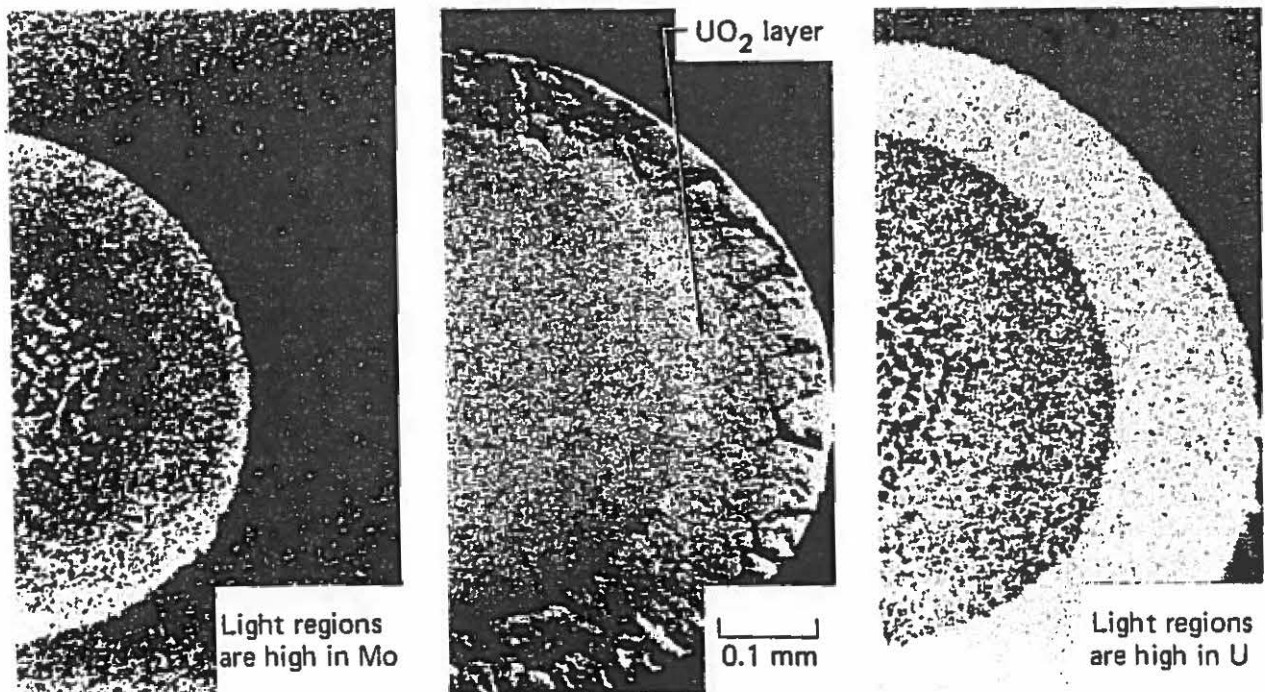UO₂ layer

0.1 mm

Light regions are high in U

Fig. 1. SEM photomicrograph and EDS analyses of a cross section of the fuel debris showing a central region of molybdenum and uranium surrounded by a layer of UO₂ (WNRE).
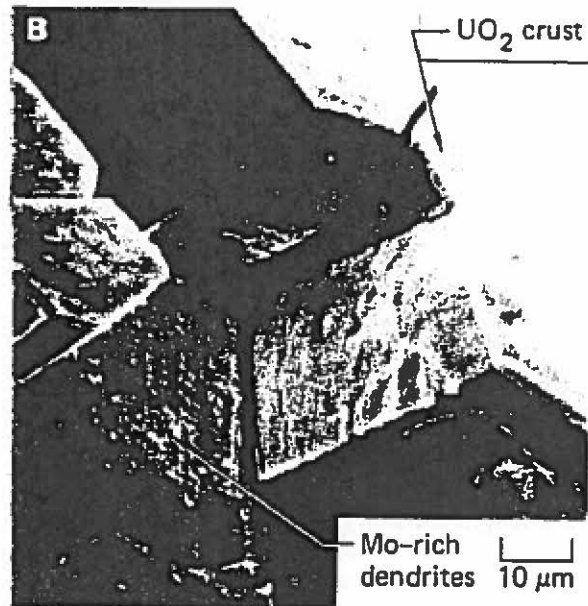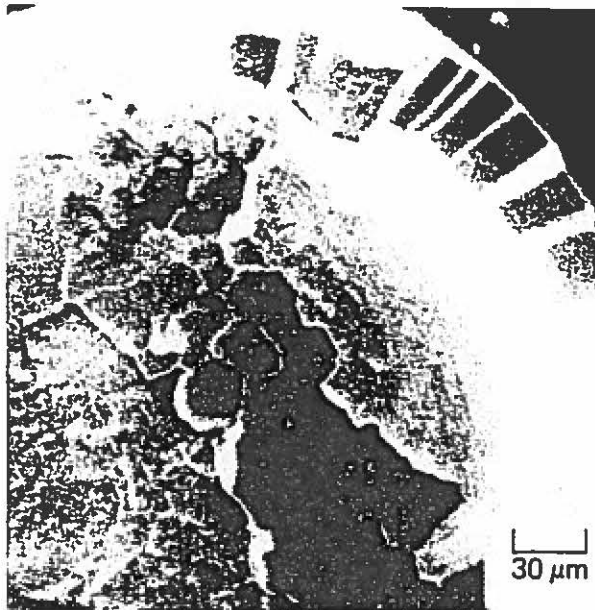
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Fig. 2. SEM photomicrographs of the debris surface showing the UO_2 crust (WNRE). Molybdenum-rich region is shown in Fig. 3.

uranium and molybdenum. Workers at WNRE postulated that the microspheres were formed from melted U-Mo alloy above 2200°C (the upper liquidus boundary) and formed the UO_2 crust during reentry. Early guesses with limited data also suggested a U-Mo alloy core or UO_2-Mo or UC-Mo cermet cores. Since positive identification of the fuel debris was not possible with techniques available at WNRE, we proposed a series of simulation studies to identify the original fuel and perhaps reveal the environment this fuel was exposed to during reactor breakup and reentry.
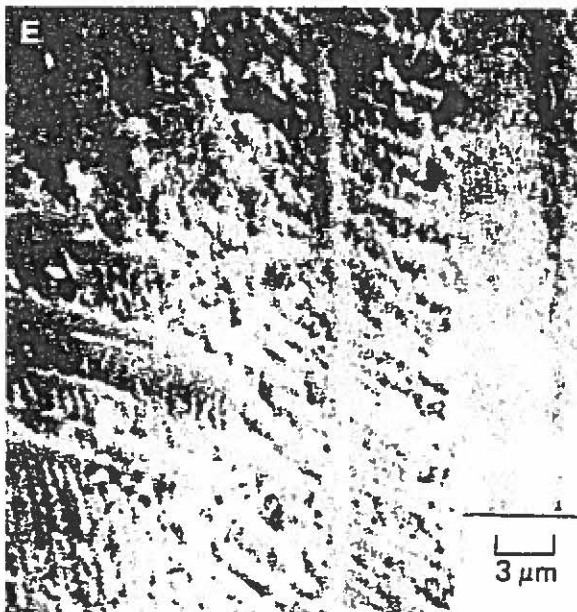
Fig. 3. SEM photomicrograph showing the molybdenum-rich dendrites, which were a prominent feature of the central region of the debris microspheres (WNRE).

UO_2-Mo Simulation

Simulation studies were made on the UO_2-Mo system to confirm or deny the possible reaction of UO_2 with molybdenum under conditions expected during reactor breakup and/or reentry. Reduction of UO_2 to uranium and volatilization of some molybdenum as oxides could produce the dendrites observed in the microspherical debris at WNRE.

Vacuum heat treatment of four powder mixtures showed no reaction between UO_2 and molybdenum (Fig. 4), confirming earlier reports[12] of no reactivity at 2155 °C. These results are also supported by a report that no reaction was observed at the UO_2-Mo interfaces during fabrication of such cermet fuels.[13] In addition, the heat-treated compacts in no way resemble the fuel debris, which shows extensive mixing and reaction between phase and elemental components (Fig. 5).

There was a remote possibility that structures similar to those found in the recovered fuel debris could have formed in U-O-Mo or U-N-Mo systems under conditions of extreme temperature (>2200°C) in low oxygen partial pressures. However, the values for the free energy of formation indicate that UO_2 is stable. Further evidence was obtained when an arc-melted specimen of UO_2-28 wt% Mo showed no reaction (Fig. 6). These findings seem to rule out the possibility of UO_2-Mo fuel in Cosmos 954.

U-Mo Simulations

Disagreements persist[14-16] about the equilibrium binary phase diagram for U-Mo. However, these disagreements have no effect on our interpretation of results because the disputed area is not
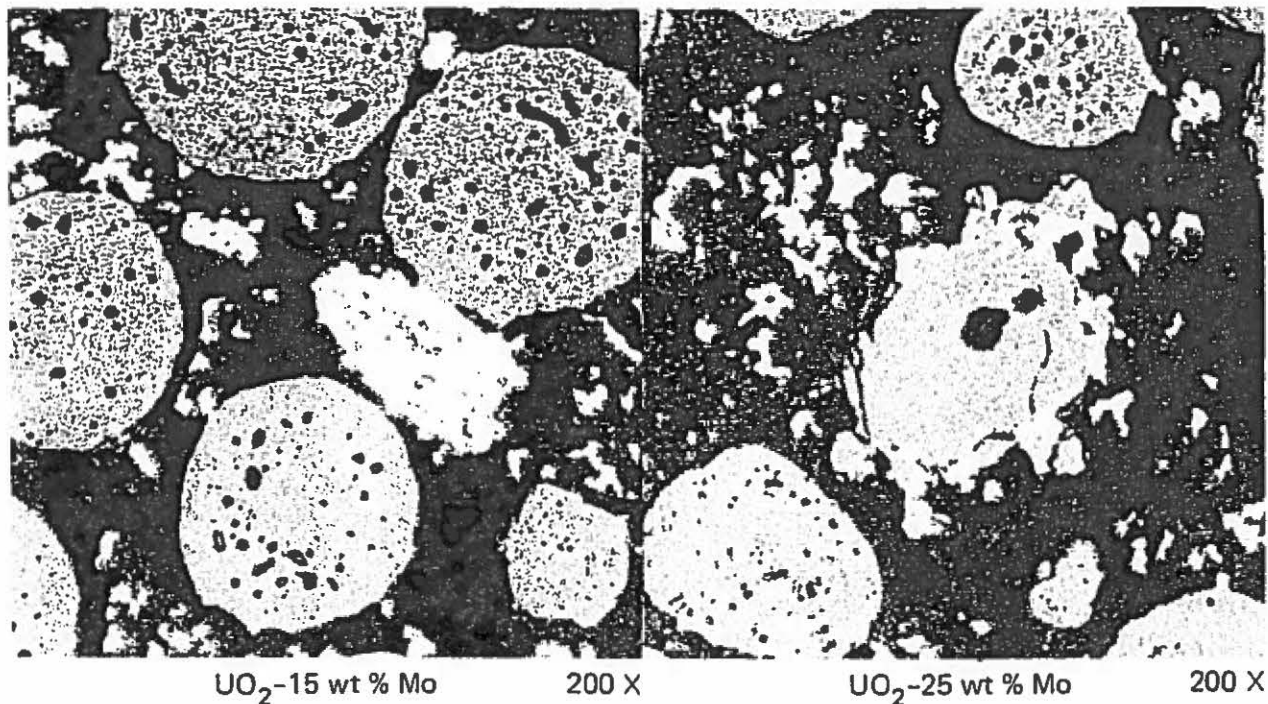
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UO₂-15 wt % Mo   200 X   UO₂-25 wt % Mo   200 X

Fig. 4. Photomicrographs of UO₂-Mo compacts heat treated at 2200°C for 40 min. Examination of the interfaces revealed no reaction between the UO₂ spheres and molybdenum. The products were porous and fragile and required epoxy infiltration bonding before cross sectioning for metallographic and x-ray diffraction evaluation. (200×).

in the region of our interest, and the debris was not formed under equilibrium conditions. The nonequilibrium state of the debris is apparent in Fig. 7.

U-8 wt% Mo and U-15 wt% Mo

Attempts to prepare U-8 wt% Mo and -15 wt% Mo specimens showed that it was difficult to prepare homogeneous samples with a molybdenum content greater than 8 wt%. Vacuum heat treatment at 1300°C for 40 min revealed that complete alloying of the molybdenum foil occurred with 8 wt% molybdenum, but not with 15 wt% molybdenum (see Fig. 8). This was interesting in itself because some microspheres had been found to contain >25 wt% molybdenum. Our experiments indicate that the high molybdenum content of the microspheres probably resulted from vaporization of uranium during breakup.

Light regions are high in Mo   SEM   20μm   Light regions are high in U
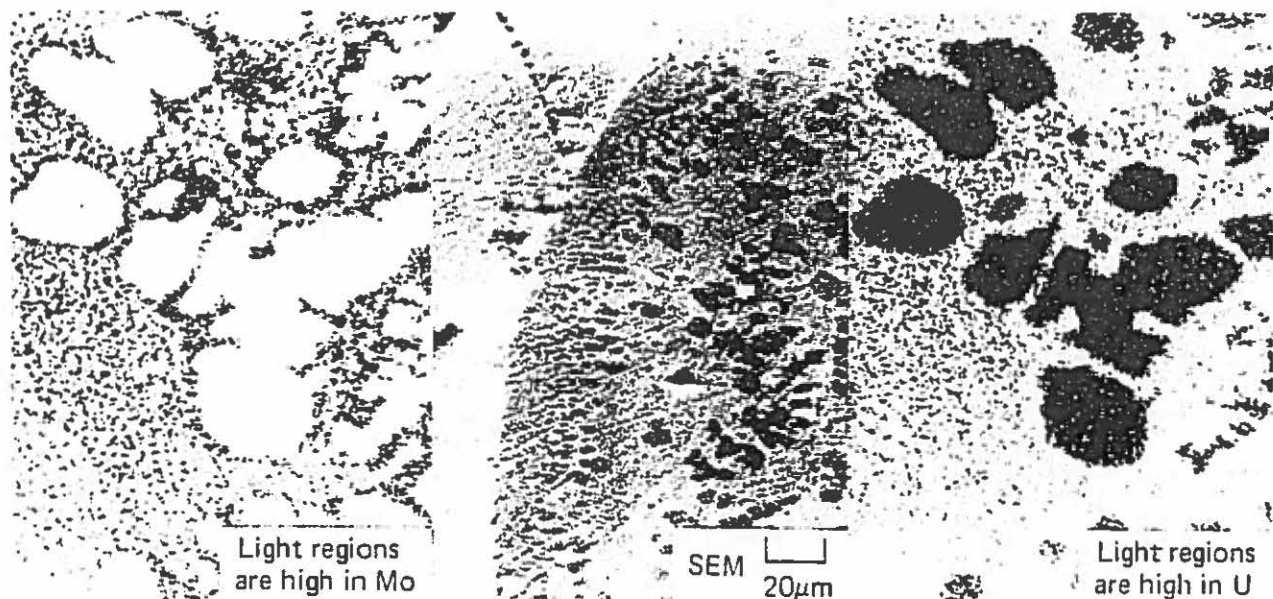
Fig. 5. SEM photomicrographs and EDS analyses of the dendritic regions in cross sections of the fuel debris. These photographs show extensive mixing of the components (WNRE).

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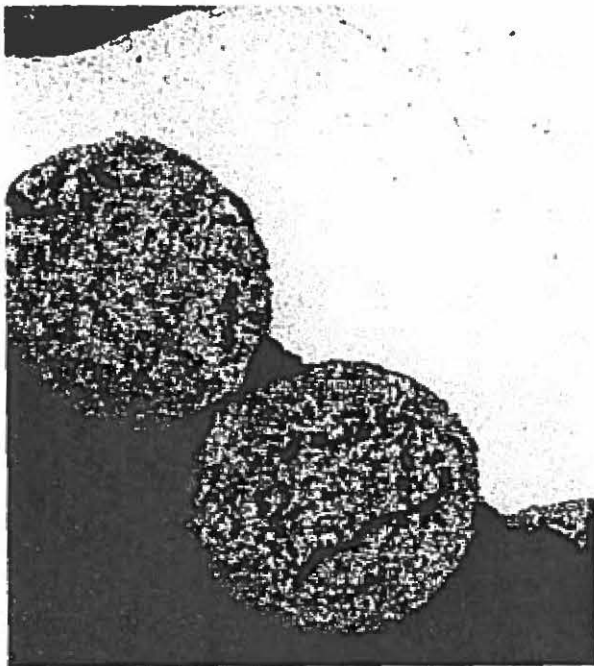
Fig. 6. Molybdenum did not react with UO₂ when arc melted on a copper chill block. This photomicrograph shows UO₂ spheres floating on the molten molybdenum. (200×).

Variations in the composition within the dendrite structure also indicate something about the history of the reactor debris. The dendrite structure in the U-15 wt% Mo button showed little variation in the U/Mo ratio; the dark regions are roughly the same composition as the light areas. Similar analyses at WNRE on the fuel debris showed significant variations in composition within the dendrite structure. We believe that these differences in composition indicate that the fuel debris cooled more slowly than these simulation samples, a logical conclusion in light of the environment (low heat transfer) in which the debris was formed.

According to x-ray diffraction, the surfaces of the 8 and 15 wt% molybdenum VHT buttons had been oxidized to UO₂ and UO (see Table I). No molybdenum was detected in the oxidation layers although they were very thin. These two-phase (UO₂ + UO) dense uranium oxide films form rapidly at 1300°C in vacuum (10 mPa) at the surfaces of the molten U-Mo alloys. The UO phase is not stable at higher temperatures and was not detected on the surface of the debris.

A button of U-15 wt% Mo was arc melted to get complete reaction and rapid quenching on the bottom of the button. The button was sectioned to examine the top, middle, and bottom. Each region had a different microstructure, and EDS analysis in the scan mode showed higher molybdenum content in the center and near the bottom (Fig. 9). The microstructure at the center of this button formed

because of the slower cooling rate at the center. This region had a much higher molybdenum content than either the top or bottom regions (Fig. 9(a) and (b)). Analysis of the two phases in the center indicated that one phase was nearly all molybdenum and the other phase was a uranium-rich alloy (see Fig. 9(c)). Segregation in the dendritic region was typical of the actual fuel debris (Fig. 10).

Differences in molybdenum content in the dendrites suggest a slow development of the dendrites. This could be explained by the debris being molten for at least 15 to 45 min at 1600°C or the debris being at very high temperatures for a short time (<15 min). Additional studies could narrow these time versus temperature relationships to give us a better understanding of reactor breakup.

U-25 wt% Mo

Vacuum heat treatments with the U-25 wt% Mo compositions showed a slow reaction with the molybdenum foil at 1300°C for 45 min. We attempted to speed up this reaction by heating to higher temperatures (1600°C) without vaporizing uranium. Nearly half of the molybdenum foil was reacted at 1600°C as compared to one third at 1300°C. After the 1600°C VHT, the larger dendritic microstructure appeared.

The surfaces of the VHT specimens of U-25 wt% Mo were oxidized to almost the same extent as the 8 and 15 wt% Mo specimens. X-ray diffraction detected only the high-temperature phase (UO₂) on the surfaces of specimens VHT at 1600°C. Two phases (UO₂ and UO) were detected on specimens VHT at 1300°C (see Table I). Only UO₂ was detected on the surfaces of the fuel debris analyzed by x-ray diffraction at WNRE. These oxide phase identifications also indicate that reactor breakup occurred ⩾1600°C.

An arc-melted button of U-25 wt% Mo was prepared to compare with the U-15 wt% Mo arc-melted button described earlier and the fuel debris. Large variations in the microstructure were again observed by metallographic examination of the button's cross section. The center of the cross section had a higher molybdenum content and there were large differences in molybdenum content between the phases. As expected, a higher molybdenum content was also detected with x-ray diffraction on this polished surface in comparison to the 15 wt% Mo arc-melted button. Again, slow cooling from above 1600°C forms microstructures similar to the debris.

The thermal stability and oxidation behavior of this arc-melted button was evaluated by a series of vacuum heat treatments at 1600°C for 1, 2, and 4 h and at 1855°C for 2 h. The three heat treatments at
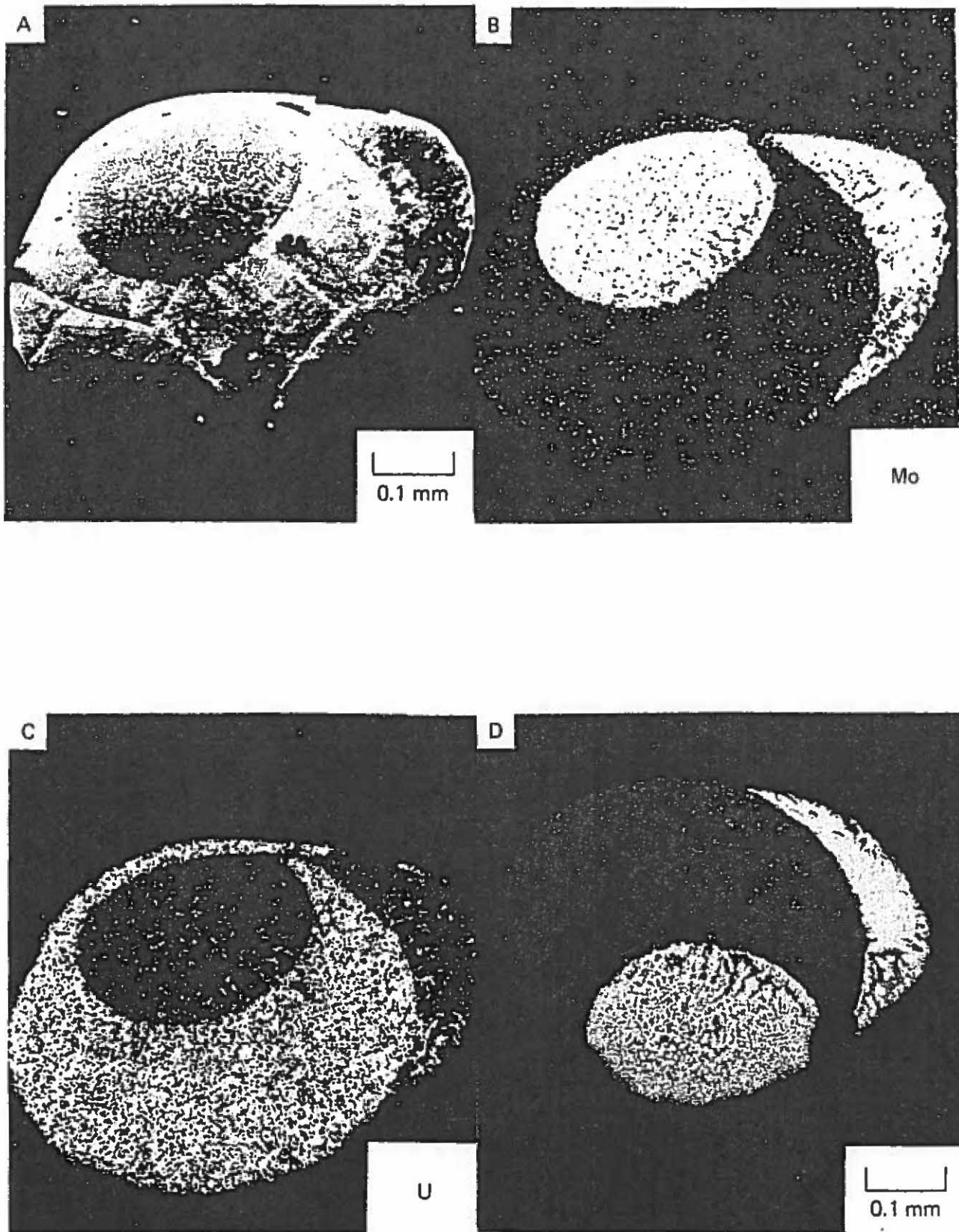
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Fig. 7. SEM and EDS examination of the cross section of a fuel debris microsphere (WNRE). The photographs show a large variation in microstructure and Mo/U ratio.
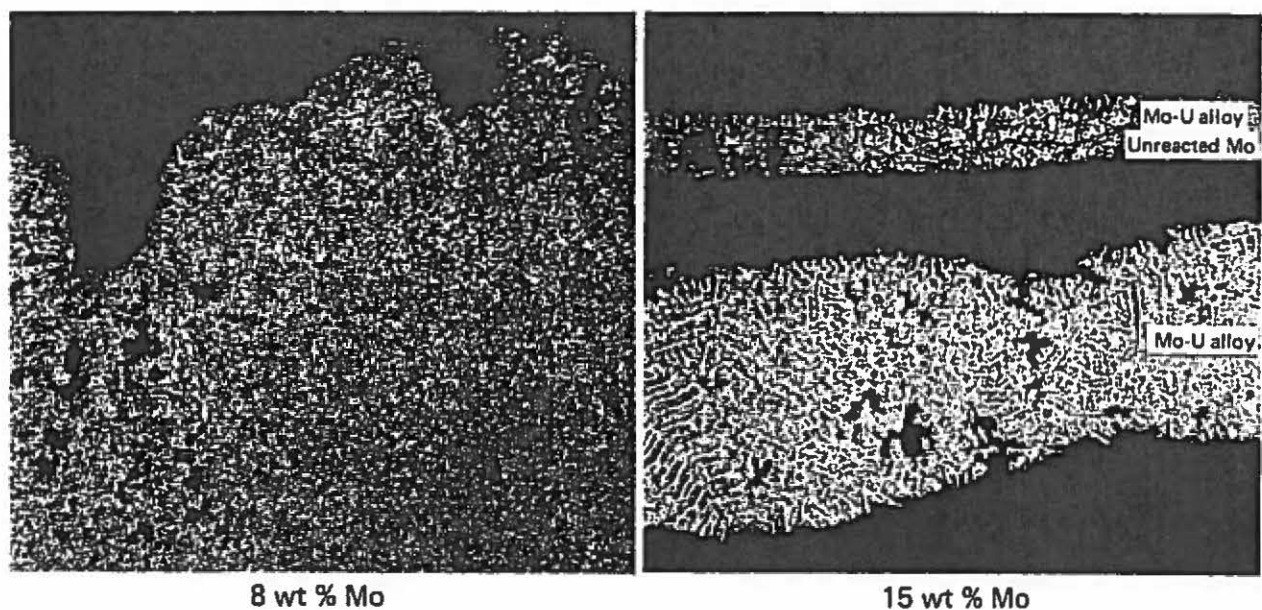
8 wt % Mo   15 wt % Mo

Fig. 8. Photomicrographs of vacuum heat treated specimens of U-8 wt% Mo and U-15 wt% Mo. Only the 8 wt% Mo mixture alloyed completely when treated 8 min at 1300°C.

1600°C had a relatively minor effect on microstructure. However, thicker oxide layers of UO_2 did form after 1 h at 1600°C. These oxide layers still contained isolated regions of U-Mo alloy. After VHT at 1855°C, a significant change in microstructure was observed (see Fig. 11(a) and (b)). The dendritic microstructure was more uniform from top to bottom, and uranium to molybdenum ratios from top to bottom were more consistent. The molybdenum-rich phase increases as the uranium in the uranium-rich phase vaporizes and/or oxidizes at the surface (see Fig. 11 b). These processes form molybdenum-rich dendritic regions similar to the regions found in the fuel debris.

While islands of U-Mo alloys and pores remain in the oxide layers on the 1600°C VHT specimens (see Fig. 12), the oxide layers on the 1855°C VHT specimen are single phase (UO_2), very dense, and contain sharp fractures (see Fig. 10(b)). These oxide layers can be compared to the UO_2 layers observed on the fuel debris in Figs. 1 and 2. Both the composition and microstructure of the high temperature oxide (1855°C) layer compare well with the UO_2 layers on the fuel debris. Sufficient time (~1 h) as

well as temperature (≥1600°C) was required to achieve oxide layers similar to the oxide layers on the fuel debris.

The type of oxide film developed on the surface of molten U-Mo alloys in a partial pressure of oxygen depends on time, temperature, and pressure. Oxidation diffusion studies of molybdenum-rich (>25 wt%) uranium alloys would help define the time-temperature-pressure conditions experienced by the fuel debris during reactor breakup.

U-40 wt% Mo

Vacuum heat treatment of the sample with the highest molybdenum content, U-40 wt% Mo, completed the developing pattern. The reaction of molten uranium with the molybdenum foil was slow and the resulting dendritic alloy had only slight variations in uranium to molybdenum ratio after VHT to 1300°C for 45 min or 1600°C for 5 min. The x-ray diffraction results in Table 1 suggest an increasing amount of molybdenum and U-Mo alloy in the uranium oxide layer at this higher initial molybdenum composition and again the absence of a UO phase at the higher temperature (1600°C). These results compare well with the previous U-Mo alloy studied. Arc-melted alloys with 40 wt% Mo were not prepared.
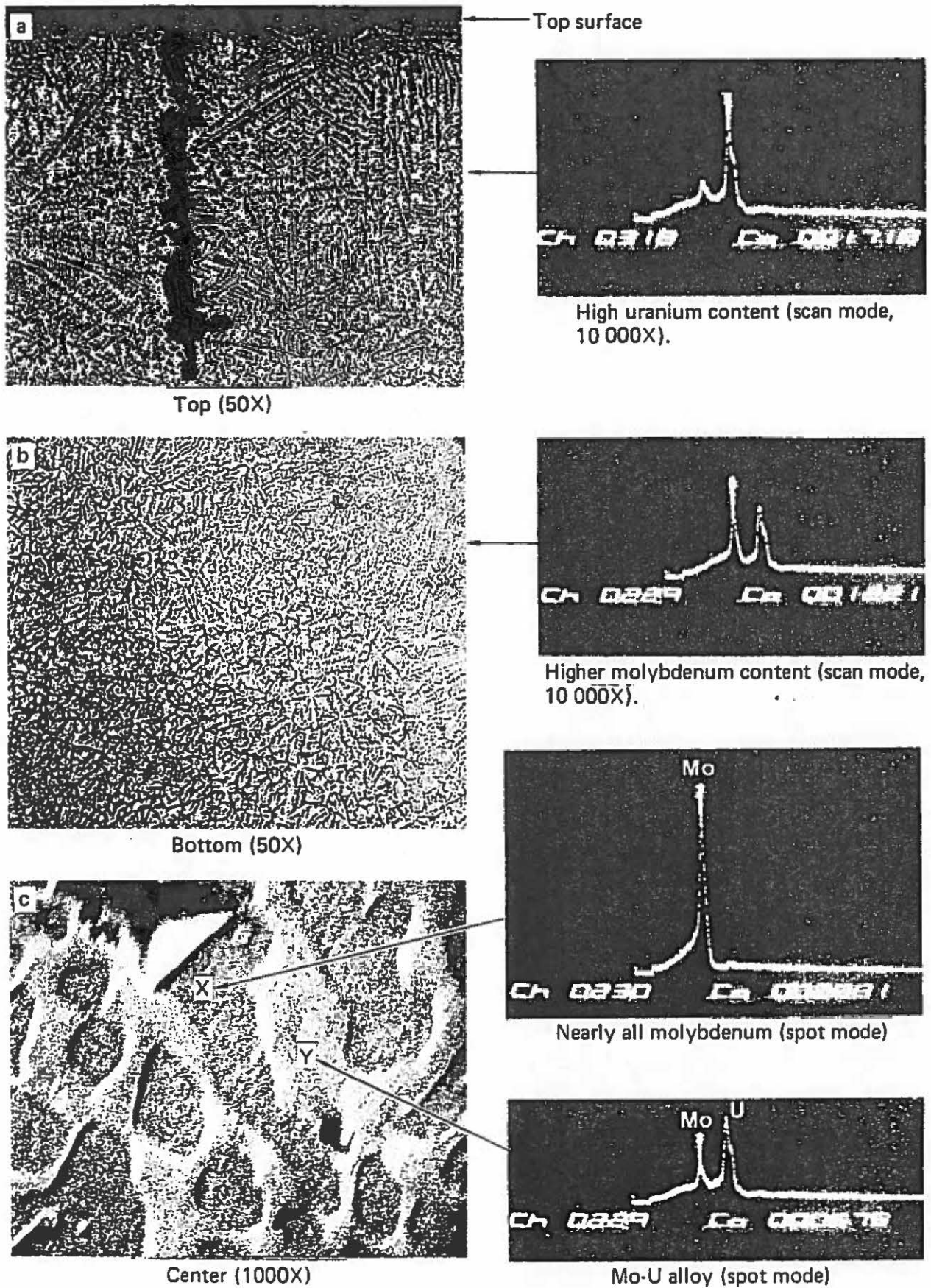
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a — Top surface

Top (50X)

High uranium content (scan mode, 10 000X).

Ch 0318   Cn 001718

b

Bottom (50X)

Higher molybdenum content (scan mode, 10 000X).

Ch 0289   Cn 001821

c

X

Y

Mo

Nearly all molybdenum (spot mode)

Ch 0330   Cn 002281

Mo U

Mo-U alloy (spot mode)

Ch 0289   Cn 002678

Center (1000X)

Fig. 9. Photomicrographs and EDS analyses of the bottom, center, and top of an arc-melted U-15 wt% Mo button. The microstructure and composition varied greatly from region to region because of differences in the cooling rate.

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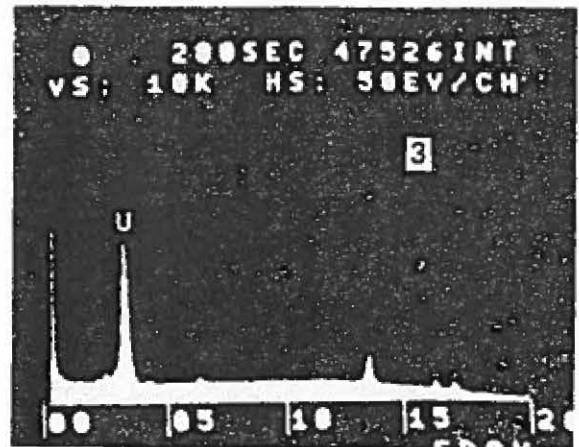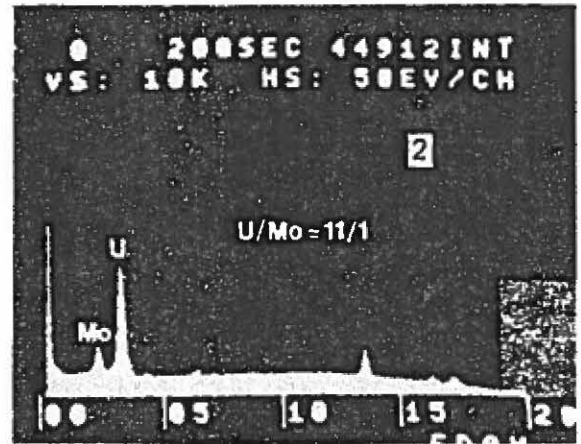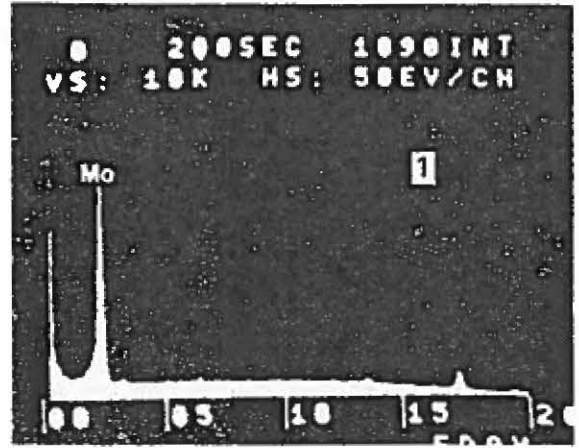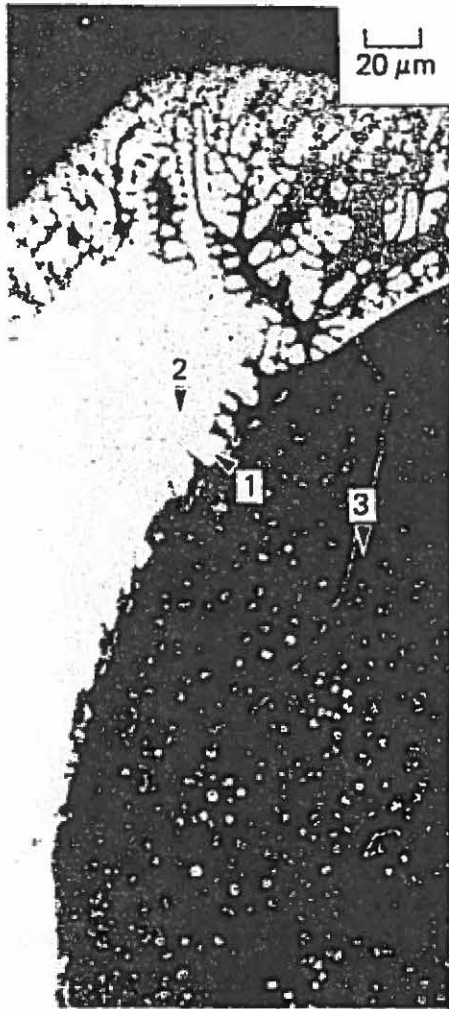
Fig. 10. WNRE analysis of the debris. Our EDS analysis also showed a great deal of segregation of uranium and molybdenum in the center of the arc-melted U-15 wt% Mo simulation specimen.

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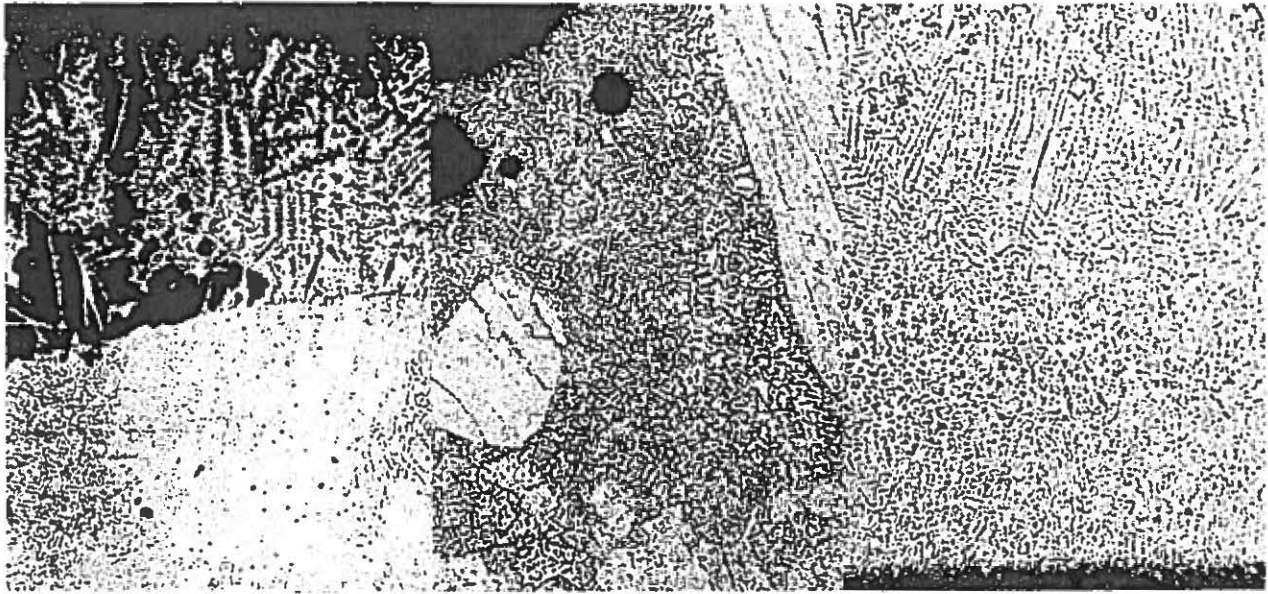
Top   Center   Bottom

(a) Arc melted button
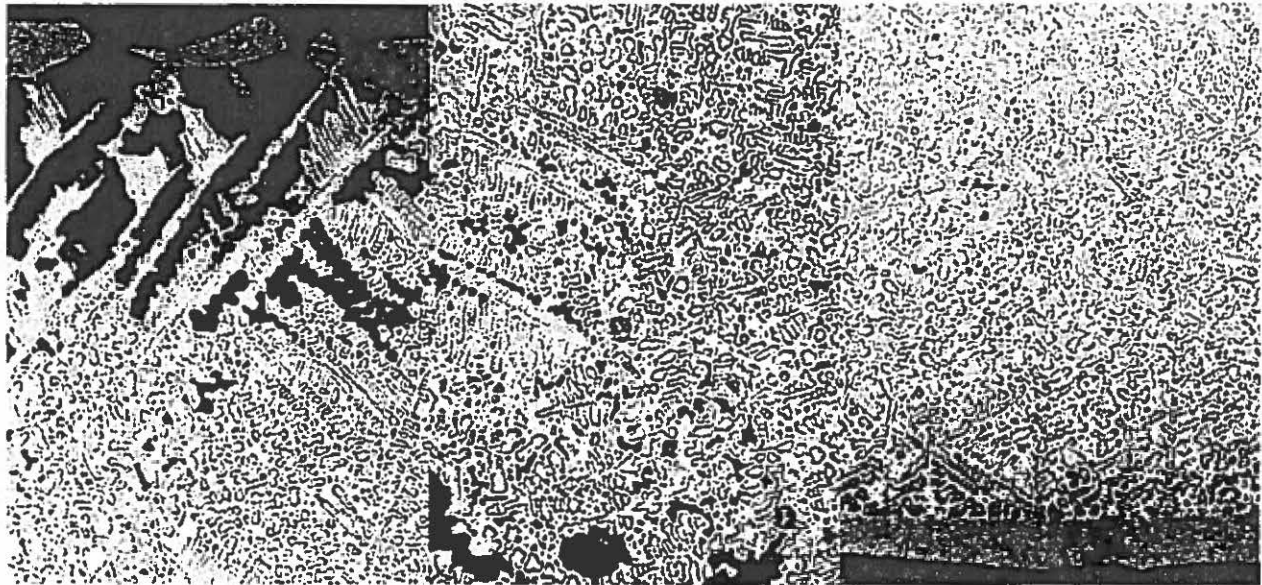
Top   Center   Bottom

(b) Arc melted button after heat treat at 1855 for 2 h

Fig. 11.   Changes in the microstructure of a U-25 wt% Mo arc-melted button after a vacuum heat treatment at 1855°C. Changes resulted from vaporization of molybdenum-rich phases and the formation of molybdenum-rich dendritic regions, which are similar to those found in the fuel debris.

12
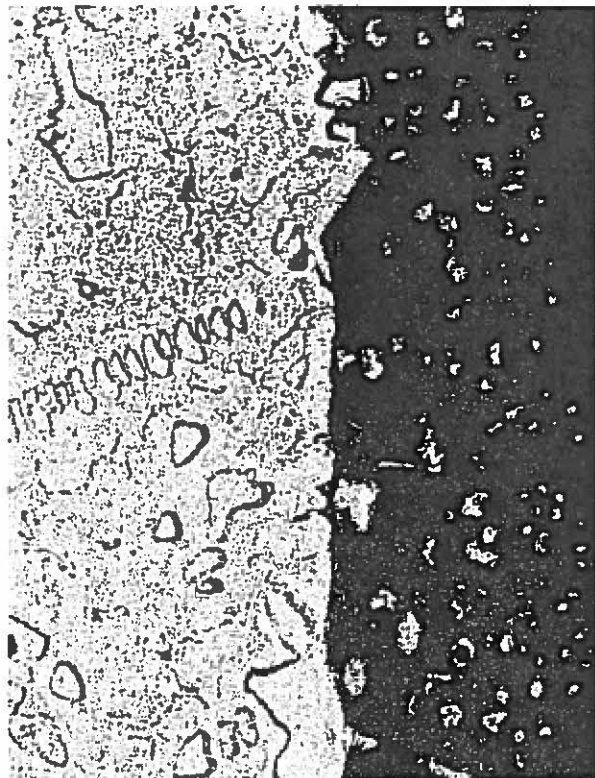
|←——Oxide layer——→|

25 wt % after 1600°C

Fig. 12. Microstructure of the oxide layer, which developed on the button that was vacuum heat treated to 1855°C.

CONCLUSIONS

The following conclusions are based on the combined results of the simulation study and results supplied by WNRE. We believe the original reactor fuel used on Cosmos 954 was U-Mo with a composition of approximately 10 wt% Mo. Additives such as Al, Si, and Fe, were originally added to the fuel to improve fabrication properties and/or phase stability. The microsphere debris was formed rapidly at 1300 to 1900°C and cooled slowly (>15 min). The surfaces of the microstructures were oxidized above 1600°C in vacuum and substantial amounts of uranium were vaporized and/or oxidized. These particles did not experience prolonged (minutes) heating at low temperatures during reentry.

ACKNOWLEDGMENTS

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HLL

APPENDIX A: CALIBRATION OF EDS WITH FOUR STANDARDS (U, UO₂, Mo, AND Nb) AT THREE ACCELERATION VOLTAGES AND TWO MODES (SPOT AND SCAN)
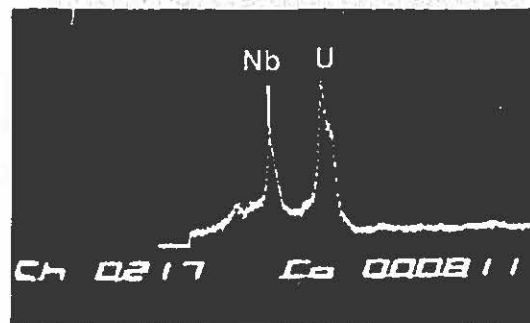
A known standard sample of uranium, UO₂, niobium, and molybdenum was used to calibrate the EDS analysis. Calibration was important because two elements (Mo and Nb) have fluorescence peaks in the same area and all three elements could have been present in the fuel. The results of the calibration are shown in Figs. A1-A6.
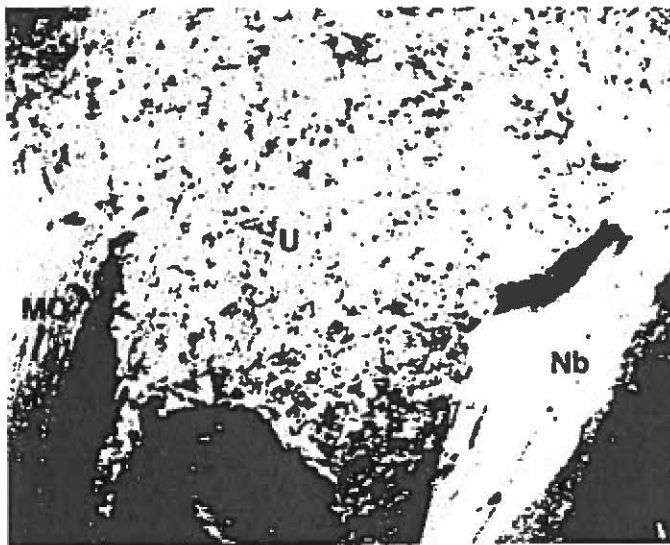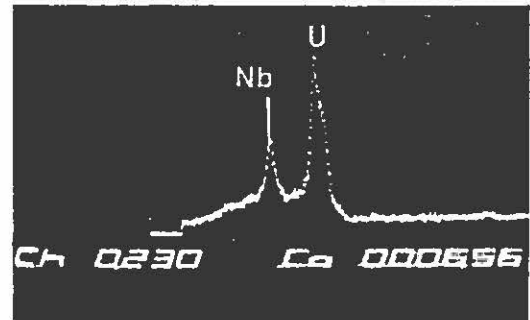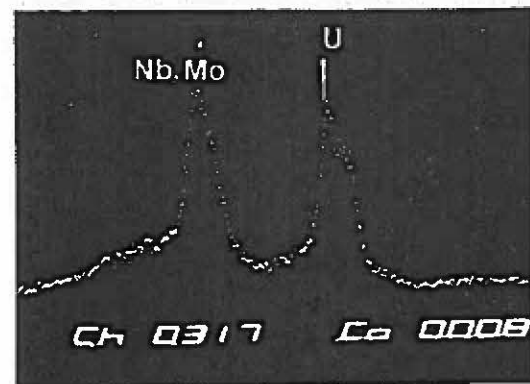

(b) Molybdenum vs uranium peak

(c) Niobium vs uranium peak

(a) Standards samples

(d) Molybdenum vs niobium peak

Fig. A1. SEM photomicrograph and EDS analyses of the U-Nb-Mo standard sample. Note the clear separation of the niobium and molybdenum peaks from the uranium peak. However, there is no resolution between molybdenum and niobium peaks. Analysis was done in the scan mode at 100×.

15

Nb   Mo

25 kV

15 kV

5 kV
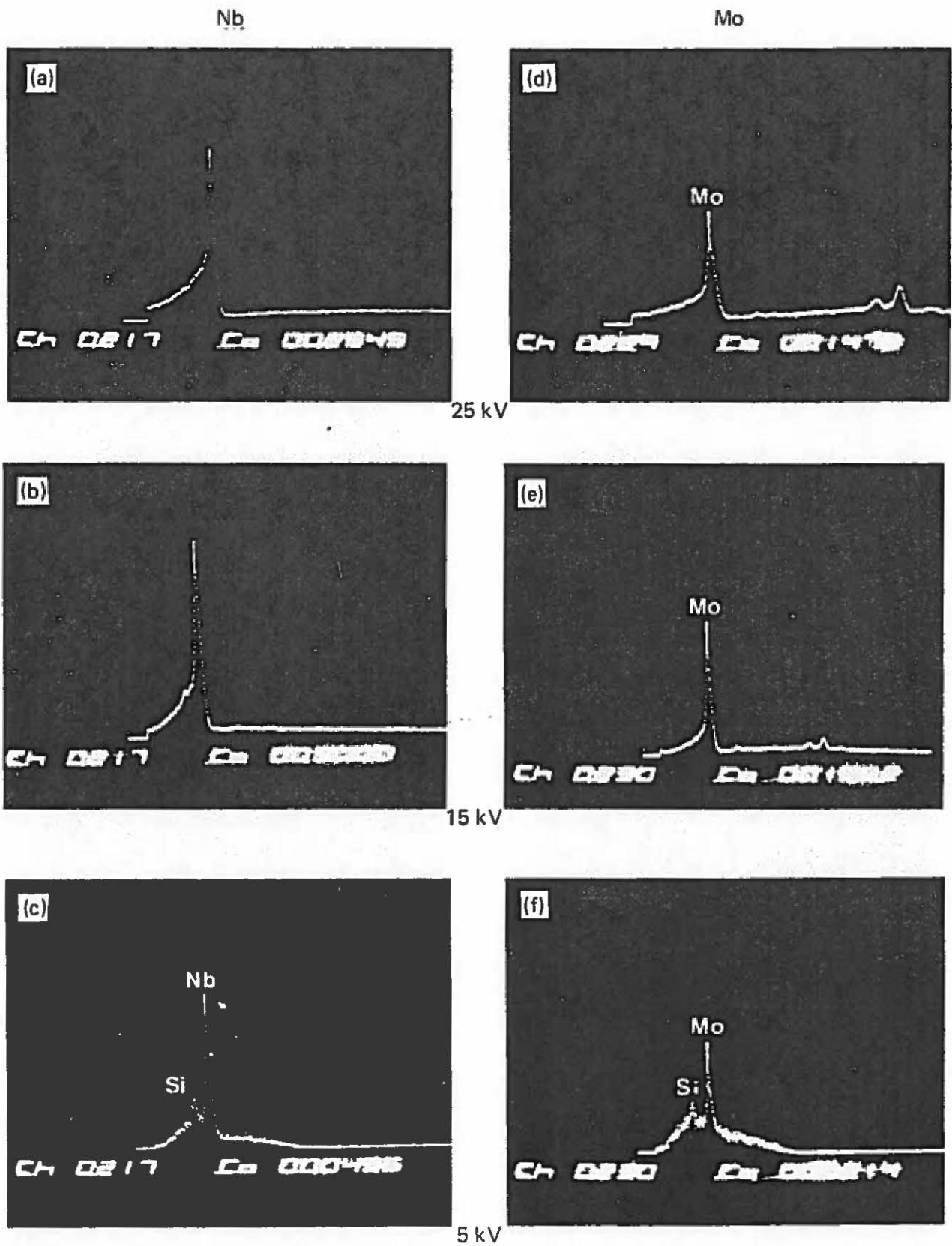
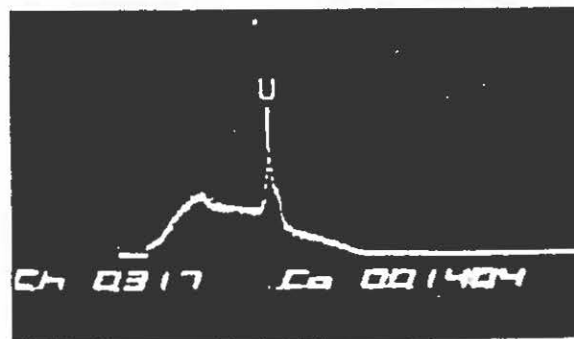Fig. A2. Detection levels of niobium and molybdenum at three power settings with scan mode (100×) on niobium and molybdenum standards.
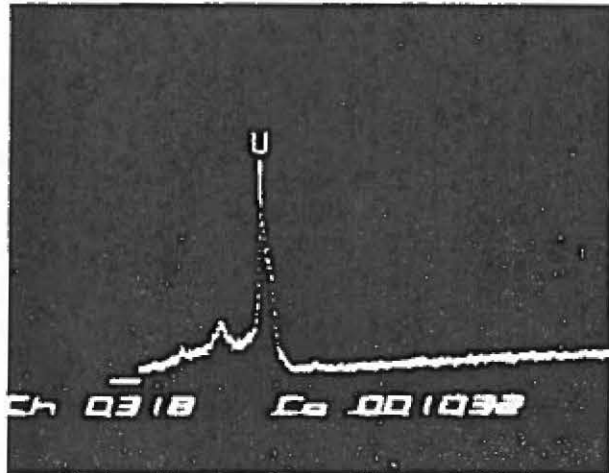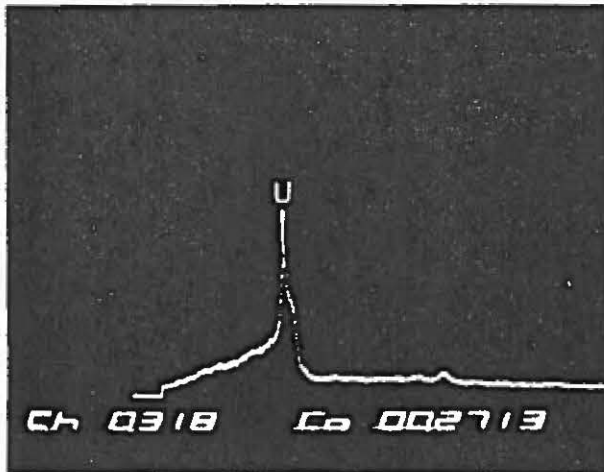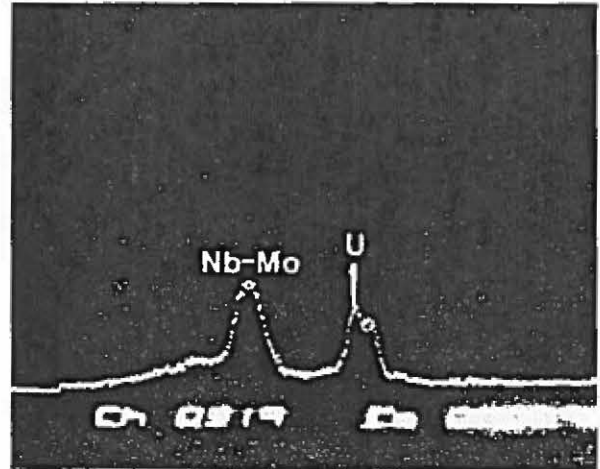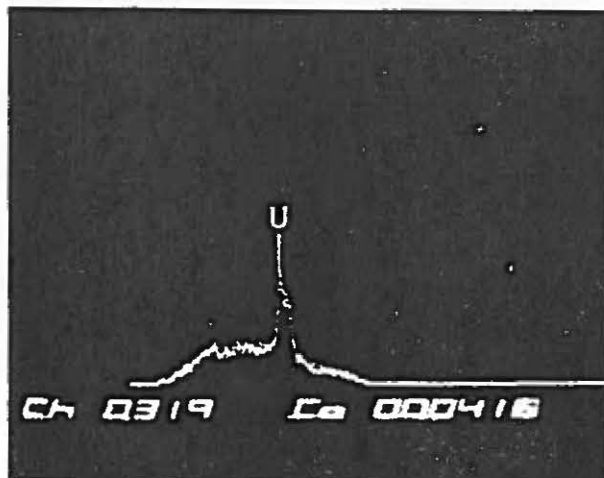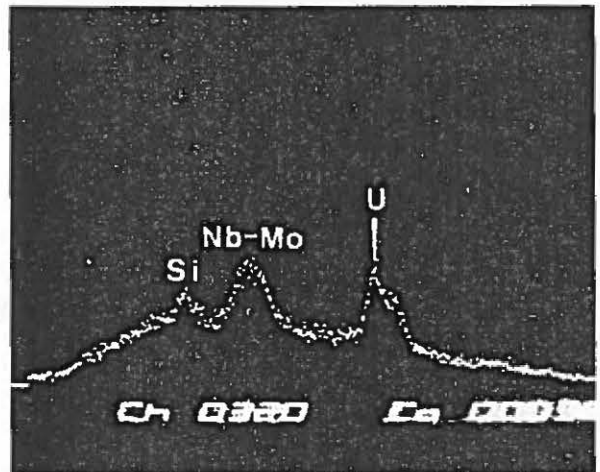
16
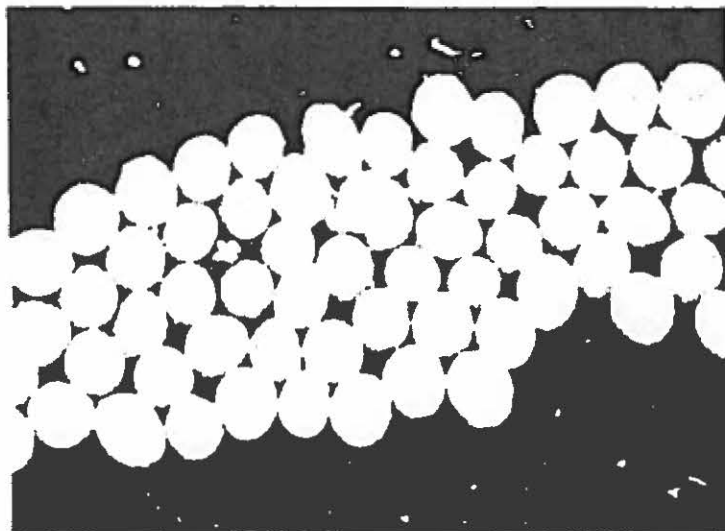
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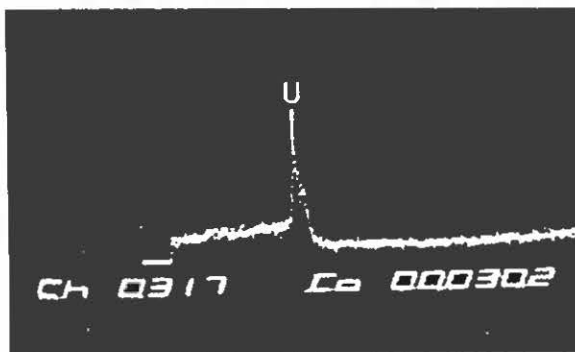
25 kV
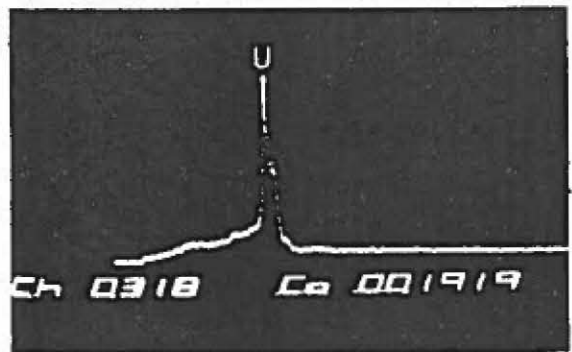
15 kV

5 kV

Fig. A3.   Detection level of uranium at three power settings with scan mode (100×) on UO_2 standard.
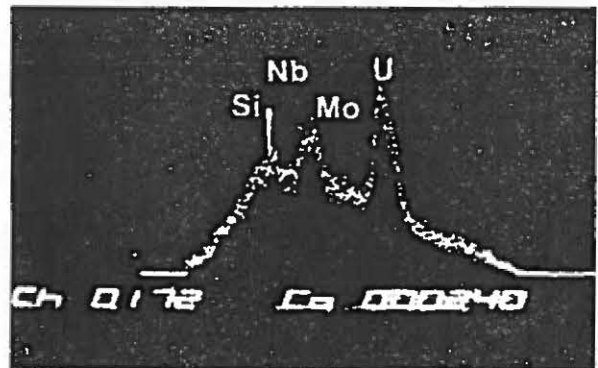
17

25 kV

15 kV

5 kV

Fig. A4. Detection levels of uranium at three power settings with scan mode (100×) on the uranium standard.

Fig. A5. Detection level of molybdenum, niobium, and uranium at two power settings and different amounts of exposed areas in scan mode on the U-Nb-Mo standard.