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N72-11536

NASA TECHNICAL MEMORANDUM

NASA TM X- 67954

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MOLYBDENUM-UO_2 CERMET IRRADIATION AT 1145 K

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TECHNICAL PAPER proposed for presentation at Winter Meeting of the American Nuclear Society
Miami Beach, Florida, October 17-21, 1971

E-6650

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SUMMARY

Two molybdenum-UO_2 cermet fuel pins were fission heated in a helium cooled loop at a temperature of 1145 K, and to a total burnup of 5.3 percent of the U^{235}. The 0.635 cm outside diameter fuel pins had a fueled length of approximately 4.57 cm and a tube wall thickness of 0.050 cm. The fueled core had 20% UO_2 with an enrichment of 93.2 percent and was fabricated by powder metallurgy technique. The fuel pin contained approximately 18 percent void. About 8 percent of this was in the form of distributed voids in the cermet. The remaining 10 percent was in the form of plenums at each end of the fuel region.

After irradiation the fuel pins were measured to check dimensional stability, punctured at the plenums to determine fission gas release, and examined metallographically to determine the effect of irradiation. Burnup was determined in several sections of the fuel pin.

The results of the post irradiation examination indicated:

(1) There was no visible change in the fuel pins on irradiation at 1145 K to 5.3% burnup.

(2) The maximum swelling of the fuel pin was less than 1 percent.

(3) There was no migration of UO_2 and no visible interaction between the molybdenum and the UO_2.

(4) Approximately 12 percent of the fission gas formed was released from the cermet cone into the gas plenum.

INTRODUCTION

The advantages of a high temperature helium cooled reactor (refs. 1, 2) have created an interest in refractory materials suitable for fuel elements.

The use of helium as a coolant permits the use of refractory metals which cannot be used in most applications because of the effects of oxidizing environments. The use of a thermal reactor (ref. 3) with its advantages of ease of control and of lower fuel inventory, dictates the use of a metal of low thermal neutron absorption cross section for the structural material in the fuel elements. Molybdenum, which has melting temperature of 2883 K (ref. 4) and a thermal capture cross section of 2.5 barns (ref. 5) and which is compatible with the UO_2 fuel (ref. 6) is a logical choice as a fuel element structural material. The low temperature ductility of molybdenum can be accommodated in the reactor design.

The gas cooled reactor designs of most interest for light weight compact reactors for surface effect vehicles are based on achieving burnups as high as possible at temperatures in the range of 1100 K to 1400 K. High burnup at this high temperature is required in order to reduce the U^{235} in-reactor inventory and avoid accidental criticality on reactor impact or melt down. Since there is no existing experimental data on the irradiation performance of molybdenum-UO_2 fuel elements at these temperatures and with high burnups, a candidate fuel geometry was tested at these conditions. The irradiation was to determine the overall performance and stability of molybdenum-UO_2 cermet fuel pins when irradiated in flowing helium at a temperature above 1100 K and to a minimum burnup of 5 percent. Significant points to be determined were whether there was (1) erosion of the molybdenum clad in the flowing helium, (2) cracking of the clad which would release fission products (3) swelling of the fuel pin which would disturb coolant flow (4) significant changes in the fuel structure which would alter heat generation profiles.

To simulate a gas cooled reactor, the tests were performed in an in-pile recirculating helium cooled loop. Two pins were irradiated with axial-flow helium coolant. In this manner the reactor design temperature and temperature profiles could be duplicated in the loop. The results of the test of the two molybdenum-UO_2 cermet fuel pins in the recirculating helium-in-pile loop are presented herein.

APPARATUS

Fuel Pins

The two fuel pins irradiated in this test were cermet pins with a molybdenum-UO_2 dispersion core, clad with molybdenum tubing. The dispersion contained 80 weight percent molybdenum and 20 weight percent of UO_2 enriched in U^{235} to 93.2 percent. The U^{235} content was 1.8 grams per pin. The specimens were made by blending the powders, hydrostatically pressing at 50,000 psi and sintering 12 hours in hydrogen at 1800 K. The UO_2 fuel had an average particle size of 50 microns. The molybdenum powder had an average particle size of 3 microns. The final molybdenum-UO_2 cermet had a density of 92 percent of theoretical. A low density was desired to allow accumulation of fission gas. The fuel cores were ground to dimensions and pressed into a honed molybdenum tube. End closure pieces were electron beam welded in place, leaving a 0.25 cm long plenum at each end. Plenum volume was 10 percent of the clad internal volume. The end pieces had a thermocouple well for measurement of the fuel pin temperature during irradiation. A cross section sketch of the fuel pin is shown in figure 1. The molybdenum conduction insulators on the fuel pin end pieces are used to minimize end heat conduction losses to the holders, which support each end of the pin during irradiation, and thus to flatten the axial temperature profile along the fuel pin during irradiation. Fuel pin end temperatures were measured with tungsten-rhenium thermocouples. Two identical pins were irradiated in parallel in the same loop.

Irradiation Facility

The irradiation described in this report was performed in a specially designed helium cooled recirculating loop (ref. 7) (fig. 2). The loop was designed to be self contained with the vane type helium pump and its electric motor drive contained in the same tube as the fuel pins being tested.

Recirculation of the helium was achieved by using concentric tubing. The flow traveled from the pump down the center tube to the fuel pins, which are mounted in parallel tubular flow channels, and back to the pump through the annular region. The annular space between the concentric tubes allowed cooling of the helium on its return flow to the pump after it had cooled the fuel pins. The fuel pins were held concentric in the cooling channels by holders which supported 0.3 cm of each fuel pin end.

The loop was of sufficient length to allow insertion of the fuel pins into the desired flux while the motor and rotating vane pump were sufficiently far from the core so that they would be in a low gamma and neutron region. During test the entire loop is sealed in a test hole which is cooled with flowing water. Thermocouples, motor power, and helium gas feed and bleed lines penetrate the best hole seal.

Helium Coolant

High purity helium was used in the test, since the molybdenum fuel pin structure was subject to attack by impurities in the gas coolant. The analysis of the helium is given in table I. In order to maintain the purity of the helium used in the test, the testing facility was pumped down before the test and purged with purified helium. During operation the helium was continously bled from the test loop at a rate of 10 cc/minute. The helium bleed served to indicate any possible fuel pin rupture by the appearance of fission products in the bleed gas. The helium bleed line was continously monitored with a radiation detector.

PROCEDURE

Irradiation

Prior to the capsule startup, the helium coolant mass-flow rate was adjusted so that the fuel pins would be over cooled when the capsule reached full power. After full capsule power was reached the desired pin clad temperature was secured by reducing the helium coolant mass flow by either changing motor RPM or by changing the perssure of the helium.

Post Irradiation Examination

The fuel pins were examined as they were removed from the loop in order to detect any gross changes in appearance. The fuel pins were measured and weighed. One pin was punctured by drilling into an end plenum in order to collect fission gas. The fuel pin was sectioned in seven pieces of approximately equal length fuel sections (fig. 1). Pieces 2, 4, and 6 were used for burnup determination and pieces 1, 3, 5 and 7 were sectioned for metallographic samples. The samples were ground through 600 grit and polished with Linde B.

RESULTS AND DISCUSSION

Fuel Pin Measurements

Visual examination of the two fuel pins as they were removed from the irradiation loop showed no change in appearance of the pins. A photograph of an irradiated pin is shown in figure 3.

Fission heating the fuel pins in circulating helium to high burnup had no apparent adverse effect on the overall integrity of the molybdenum clad. In neither fuel pin was there any indication of errosion, cracking, blistering of the clad, or warping of the fuel pins. However, dimensional measurement of the outside diameter of the clad revealed a 0.005 cm increase in diameter. This increase is less than 1 percent. The results of the pre-irradiation and post-irradiation measurements of the fuel pins are shown in table II, Part I. The measurements indicate a slight but measurable swelling of the fuel pins.

The diameter of the irradiated core was measured and compared with the pre-irradiated diameter. The core diameter also increased by 0.005 cm (table II, Part II), so the growth of the fuel pin is due to swelling of the cermet core at the conditions of the irradiation.

The average burnup determined by the analysis of fission produced Cs^{137} was 5.3 percent of the U^{235}. In addition to the Cs^{137} burnup analysis, the burnup was also measured by the mass spectrometric determination of change in uranium isotope ratios. The burnup determined by uranium isotope analysis was 5.9 percent. The mass spectrometric analysis of the uranium isotope serves as a check on the Cs^{137} analysis. The burnup analysis is given in table III. No significant variation in burnup occurred along the length of the fuel pin.

Post Irradiation Fuel Characteristics

Photomicrographs of a transverse section of the irradiated fuel core are shown in figures 4(a) (100x polished) 4(c) (100x etched) 4(e) (250x polished) and 4(g) (250 x etched). The as fabricated unirradiated fuel core is shown in a similar set of figures in 4(b), (d), (f), and (h).

The smaller black areas in the molybdenum matrix are voids which resulted from using a sintering temperature which would produce a cermet core of only 92.5 percent density. The voids are either small spherical holes or irregular pockets resulting from the void collecting at grain boundaries during sintering. There are also a few UO_2 pull outs, resulting from dislodging of UO_2 during metallographic preparation. These can be distinguished from the porosity by their size, which is approximately that of the UO_2, and by their shape, which is roughly spherical. The small round holes in the UO_2 are voids approximately 1 micron in diameter.

From inspection of the photomicrographs it is evident that during this irradiation at 1145 K to 5.3 percent burnup there is no visible change in the structure or the amount of the porosity of the molybdenum matrix or of the UO_2 fuel. There is no visible change in the density of the UO_2. There is no migration of the UO_2 along grain boundaries of the molybdenum matrix. The fuel after irradiation to 5.3 percent burnup at 1145 K shows no significant change from the pre-irradiation structure.

Fission Gas Measurement

In order to contain the fission gas produced in the high burnup scheduled for this pin, the molybdenum-UO_2 cermet fuel core was sintered to only 92 percent of theoretical density. It was anticipated that the porosity in the cermet fuel core would hold the fission gas which might be released from the UO_2 at the temperature and burnup of the experiment. The volume of fission gas formation per pin for 5.3 percent burnup of the U^{235} is 2.6 cc at STP.

One of the irradiated fuel pins was punctured and the volume of fission gas released to the plenums was measured. The volume of fission gas was 0.29 cc at STP. This represents a release of only 12 percent of the total fission gas formed. While it might be expected that at this burnup and temperature of irradiation that a large percent of the gaseous fission products formed would escaped from the UO_2, either the 8 percent porosity built into the cermet core trapped the released fission gases or they were retained in the UO_2 or by the molybdenum surrounding the UO_2 which acted like small pressure vessels.

In order to determine the distribution of fission products, two autoradiographs were made of the transverse sections of the irradiated cermet core. The results are shown in figure 5(a) and (b). The even distribution indicates that there was no significant migration of fission products. The autoradiograph in 5(a) was metallographic sample 3 and that in 5(b) was metallographic sample 5. In addition to the absence of a transverse gradient of fission products, the results indicate there was no longitudinal migration of fission products. It is apparent that at the temperature of the irradiation most of the fission products are locked in the cermet.

SUMMARY OF RESULTS

Two molybdenum UO_2 cermet fuel pins with molybdenum clad were fission heated in a forced convection helium coolant for sufficient time to achieve 5.3 percent burnup. The cermet core contained 20 weight percent of 93.2 percent enriched UO_2. The results of theinvestigation were as follows:

(1) There was no visible change in the appearance of the molybdenum clad during irradiation.

(2) The maximum increase in diameter of the fuel pins was 0.8 percent.

(3) There was no migration of the UO_2 along grain boundaries and there was no evident interaction between the molybdenum and the UO_2.

(4) Approximately 12 percent of the fission gas formed was released from the cermet core into the gas plenum.

CONCLUSIONS

The results of the irradiation showed that cermet fuel pins of this design could be satisfactorily irradiated at 1145 K to over 5 percent burnup with less than 1 percent swelling. This test also showed that these fuel pins could successfully contain the fission products at this burnup and, that the cermet was resistant to migration of fuel.

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TABLE I. - COMPOSITION OF HELIUM COOLANT

Component	Analysis, ppm
N_2	<5.0
O_2	.5
Total hydrocarbons	<1.0
CO_2	<.5
CO	<1.0
CH_4	<.5
NO	<.1
C_2H_2	<.05
H_2O	.15
He	Balance

TABLE II. - PIN DIAMETER

Station	Pre irradiation (avg of 4 measurements), cm	Post irradiation (avg 4 measurements), cm	Change, cm	Change, percent
Part I - Clad				
A	0.6332	0.6358	0.0026	0.4
B	.6330	.6383	.0053	.8
Part II - Core				
	(Avg 2 measurements), cm	(Avg 2 measurements), cm		
	0.5321	0.5375	0.0054	1.0

TABLE III. - BURNUP ANALYSIS

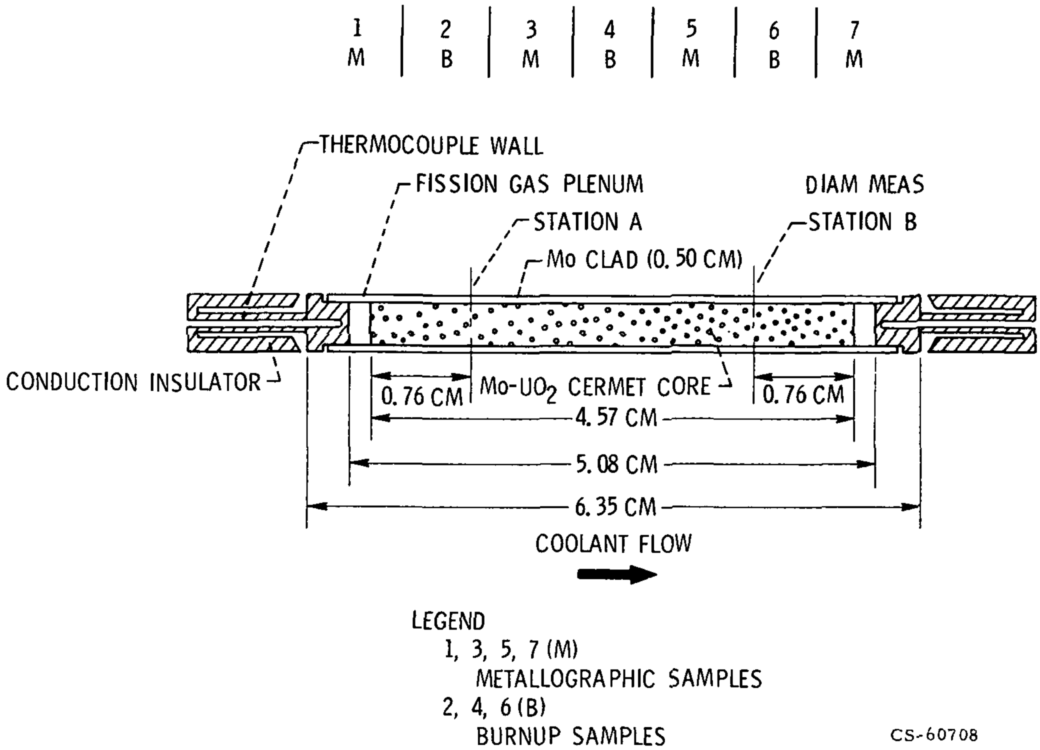
Sample	Burnup by Cs^{137}, percent	Burnup by U isotopes, percent
2B	5.53	5.98
4B	5.13	5.86
6B	5.35	5.94
Average	5.34	5.93

Note: Constants used in calculations

Half life of Cs^{137} 29.2 yr

Fission yield of Cs^{137} 6.15 percent

FUEL PIN



CS-60708

Figure 1

IRRADIATION LOOP

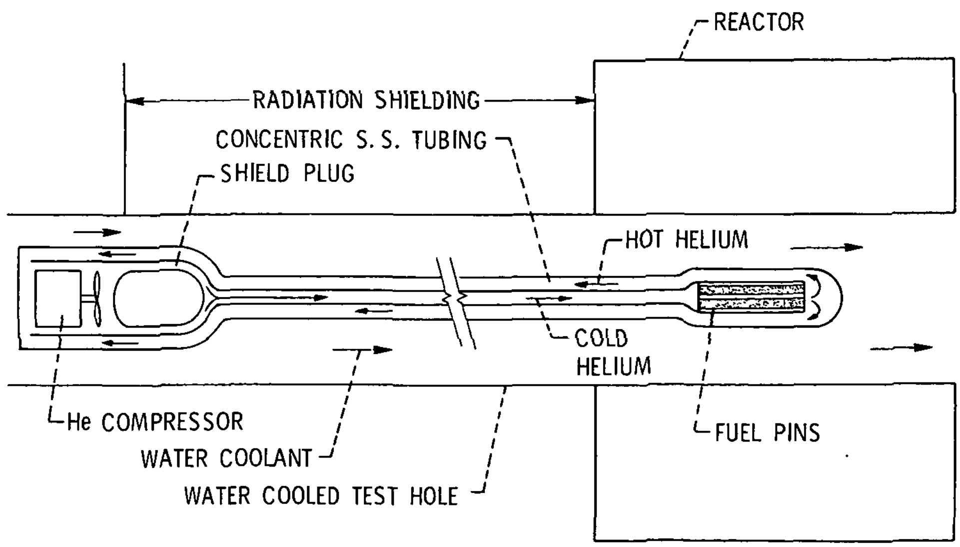


Figure 2

CS-60709

MOLYBDENUM-UO_2 FUEL PIN AFTER IRRADIATION

CONDUCTION INSULATORS REMOVED FROM ENDS

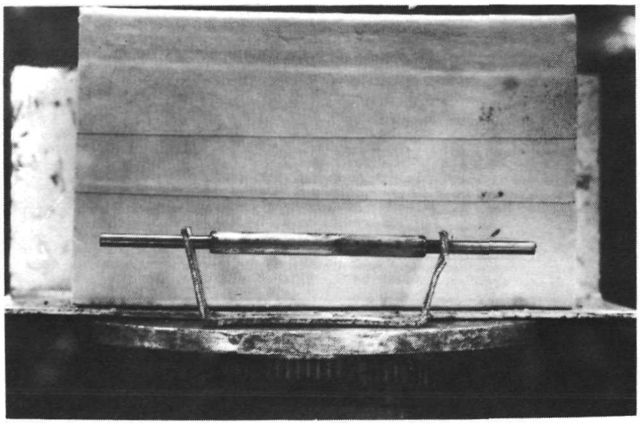


Figure 3 CS-60717

MOLYBDENUM-UO_2

IRRADIATED AS POLISHED, X100

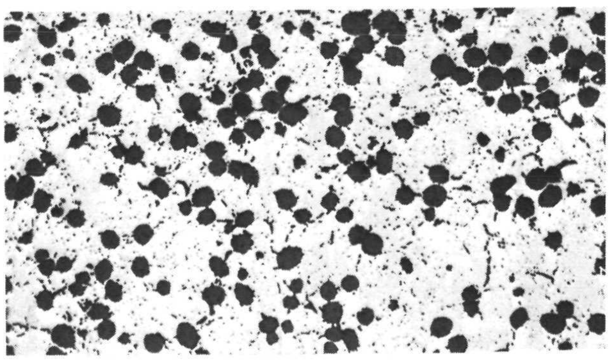


Figure 4(a) CS-60710

MOLYBDENUM-UO_2

UNIRRADIATED AS POLISHED, X100

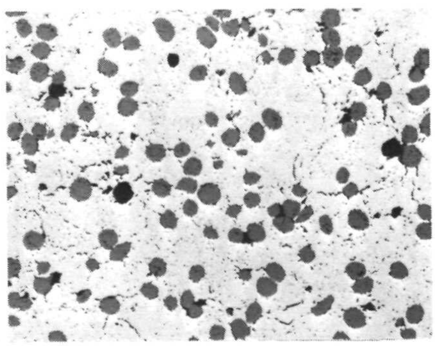
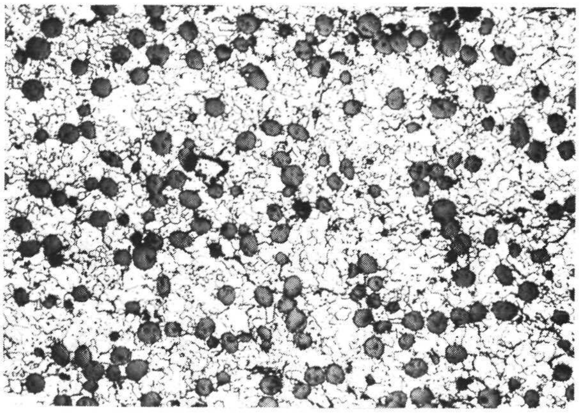


Figure 4(b) CS-60716

E-6650

MOLYBDENUM-UO_2

IRRADIATED ETCHED, X100

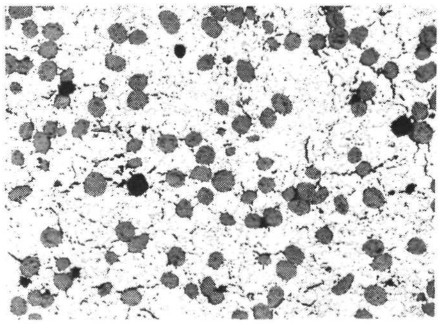


CS-60713

Figure 4(c)

MOLYBDENUM-UO_2

UNIRRADIATED ETCHED, X100

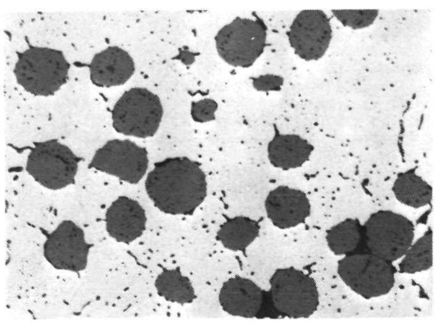


CS-60714

Figure 4(d)

MOLYBDENUM-UO_2

UNIRRADIATED AS POLISHED, X250

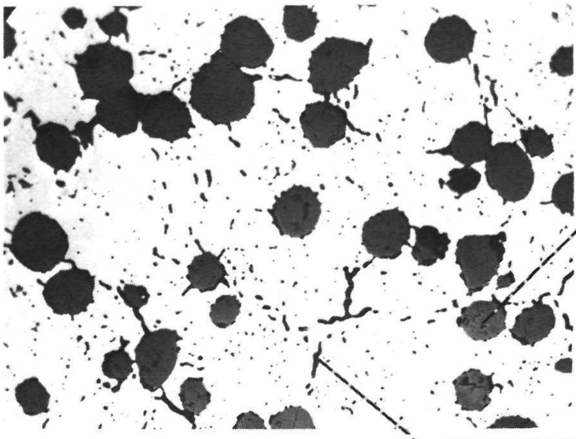


CS-60711

Figure 4(e)

MOLYBDENUM-UO_2

IRRADIATED AS POLISHED, X250

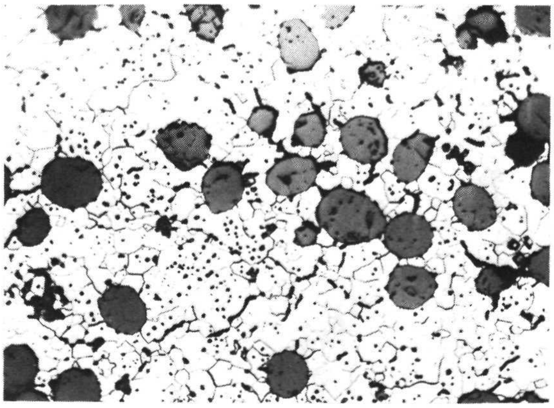


CS-60720

Figure 4(f)

MOLYBDENUM-UO_2

IRRADIATED ETCHED, X250

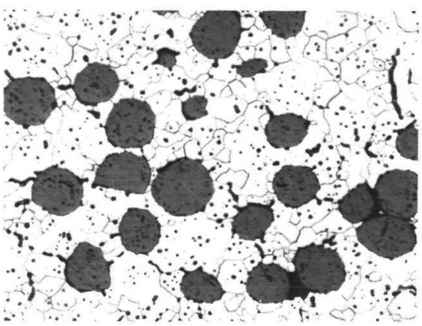


CS-60715

Figure 4(g)

MOLYBDENUM-UO_2

UNIRRADIATED ETCHED, X250



CS-60712

Figure 4(h)

E-6650

β-γ AUTO-RADIOGRAPH

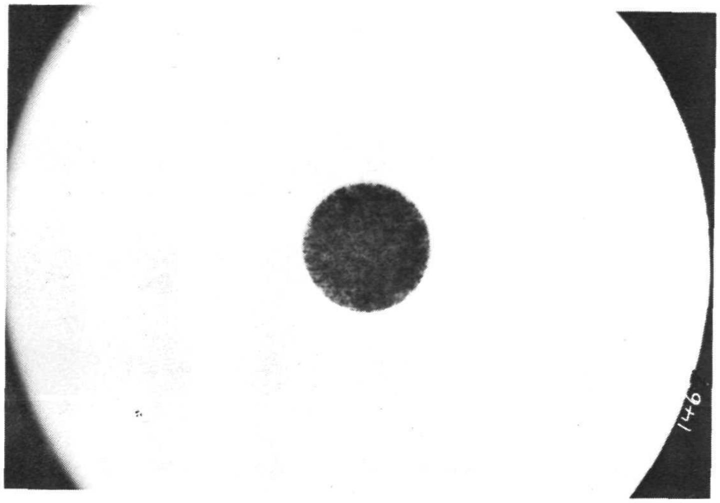


Figure 5(a) CS-60718

β-γ AUTO-RADIOGRAPH

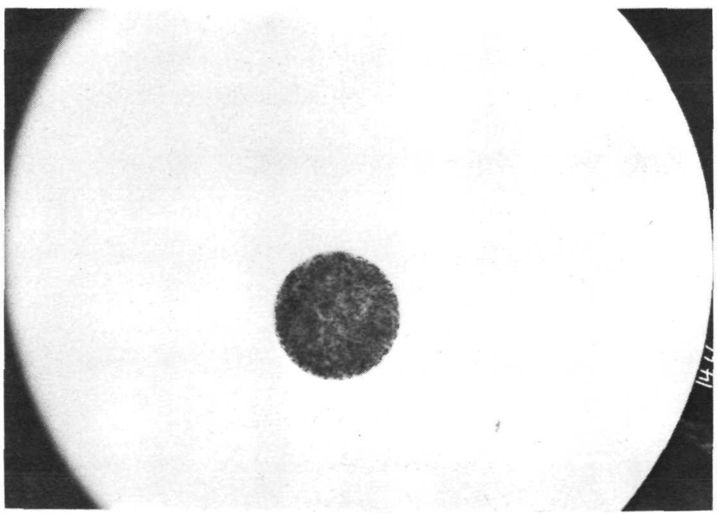


Figure 5(b) CS-60719