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CONTINUING INVESTIGATIONS FOR TECHNOLOGY ASSESSMENT OF ^{99}Mo PRODUCTION FROM LEU TARGETS

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ABSTRACT

Currently much of the world's supply of ^{99m}Tc for medical purposes is produced from ^{99}Mo derived from the fissioning of high enriched uranium (HEU). The need for ^{99m}Tc is continuing to grow, especially in developing countries, where needs and national priorities call for internal production of ^{99}Mo. This paper presents the results of our continuing studies on the effects of substituting low enriched Uranium (LEU) for HEU in targets for the production of fission product ^{99}Mo. Improvements in the electrodeposition of thin films of uranium metal are reported. These improvements continue to increase the appeal for the substitution of LEU metal for HEU oxide films in cylindrical targets. The process is effective for targets fabricated from stainless steel or hastaloy. A cost estimate for setting up the necessary equipment to electrodeposit uranium metal on cylindrical targets is reported. Further investigations on the effect of LEU substitution on processing of these targets are also reported. Substitution of uranium silicides for the uranium-aluminum alloy or uranium aluminide dispersed fuel used in other current target designs will allow the substitution of LEU for HEU in these targets with equivalent ^{99}Mo-yield per target and no change in target geometries. However, this substitution will require modifications in current processing steps due to (1) the insolubility of uranium silicides in alkaline solutions and (2) the presence of significant quantities of silicate in solution. Results to date suggest that both concerns can be handled and that substitution of LEU for HEU can be achieved.

INTRODUCTION

Technetium-^{99m} for medical purposes is a decay product of ^{99}Mo, which is produced in research reactors from the fissioning of ^{235}U or from neutron capture of ^{98}Mo. This continuing effort is related only to fission-product ^{99}Mo.[1] Presently, ^{99}Mo is produced from a variety of target designs that contain HEU (~93% ^{235}U). These designs include curved fuel plates,[2,3] rods,[4] and cylinders with a film of UO_2 electroplated on the inside surface[5,6].

The overall purpose of this study is to assess the feasibility of substituting LEU for HEU in targets for production of fission-product ^{99}Mo. The main issues that were addressed during this year's efforts were (1) optimization of conditions for electrodeposition of uranium metal to produce well-bonded, uniform, dendrite-free, 0.3mm films that are compatible with current target designs using UO_2 films, (2) further studies on the effects of LEU substitution on the processing of ^{99}Mo from these targets, and (3) the dissolution by base and separation and purification of ^{99}Mo from uranium silicide targets. This paper reports on the present status of our investigations.

FABRICATION OF LEU METAL TARGET

Several target designs with HEU are currently used for the production of fission product ^{99}Mo. These designs include curved plates[2,3] and rods[4] that contain either U-A alloy or aluminide fuel and cylinders coated with a thin film of UO_2 on the inside surface.[5,6] We anticipate that the plate and rod target designs can be converted to LEU by utilizing the high uranium densities achievable with the new LEU silicide fuels[7] without changing the target geometries. However, the cylindrical target design currently uses a UO_2 surface density with HEU that in some cases is close to its practical fabrication limit. A new target design and/or a much denser film material is required for use of LEU. As a result, our efforts have concentrated on a cylindrical design with a thin layer of uranium metal electrodeposited on the inside surface.

Uranium metal targets for fission ^{99}Mo production have several advantages over the UO_2 targets; e.g., uranium metal is about twice as dense as UO_2, its thermal conductivity is an order of magnitude higher than UO_2, and its plating efficiency from LiCl-KCl-UCl_3 molten salt melts is 100% vs. 20% for the UO_2 deposition process. The principal disadvantages of using electrodeposited uranium metal targets are that (1) they must be prepared from molten salt systems at high temperatures (~450°C) in an inert atmosphere and (2) the deposit morphology tends to be dendritic. While the higher conductivity and density make uranium metal targets appear quite promising, the quantity of ^{235}U in the uranium metal targets will be slightly higher than in HEU targets,[1] and heat management and safety problems are concerns that need to be thoroughly analyzed.

Published literature indicates that uranium metal can be electrodeposited from either a fluoride[9-13] or chloride[8,14-18] molten salts melt. Because the fluoride melts are more corrosive, more sensitive to moisture, and operated at much higher temperatures than the chloride melts, electroplating uranium metal from chloride melts offers advantages over the fluoride melts. While many different base chloride salts containing UCl_3 have been used to plate uranium metal ($BaCl_2$-$CaCl_2$-LiCl, $BaCl_2$-KCl-NaCl, $BaCl_2$-$CaCl_2$, LiCl-NaCl and LiCl-KCl), none seem to offer a significant advantage over any other. Because the LiCl-KCl eutectic is inexpensive and readily available commercially, it is the base electrolyte for our studies.

This investigation evaluated the effects of current density, plating mode (direct current vs. pulse current) and metal substrate material on the quality of the uranium deposit. Initial studies evaluated the quality of the uranium deposits that were formed under direct current plating conditions using a nickel substrate material. Pulse-plating regimes were next investigated in an effort to optimize the plating regime. In these latter studies, factorial investigations were conducted to identify a regime where dendrite-free deposits are formed. Once these studies were completed, pulse-plating electrodeposition of uranium films were carried out on a variety of substrate materials. Preliminary studies with stainless steel, carbon steel and zircaloy indicated these materials gave poorly bonded, poor quality uranium deposits; therefore the emphasis of the research shifted to nickel and copper substrate materials. Nickel and copper were chosen as potential substrate materials because they are fairly resistant to air oxidation and make excellent precoating materials for either stainless steel or zircaloy cylinders. Both materials may be electrochemically deposited on the stainless steel or zircaloy or,[19,20] in the case of nickel, by electroless plating methods.[21] Intermetallic compound formation[22] between nickel or copper and uranium should result in good adhesion of the deposit to the substrate.

Electrochemical parameters investigated in our pulse-plating studies included the effect of a nucleation pulse and surface anodization prior to uranium pulse deposition. Nucleation pulses have been proposed earlier by other investigators as a tool to enhance coating uniformity,[23] while surface anodization has been proposed as a means to generate a clean surface for deposition.[24] A cost estimate for equipment to run a full-scale electrodeposition process is provided at the end of this section.

Experimental

Uranium metal was electrodeposited from molten salt in a helium atmosphere glove box. The cathode substrates used in constant-current and later pulse-plating experiments were disk-shaped coupons of metal (zircaloy, stainless steel, carbon steel, nickel, or copper) 1.6 cm dia. and 1 cm high (total exposed surface area = 6.2 cm^2). Early pulse-current experiments were run with smaller, 0.5 cm dia. disks due to equipment limitations. The cathode was rotated at ~300 rpm for constant-current experiments and was kept stationary for pulse plating experiments.

The bottom circular face of the coupon was polished for five minutes successively with 600-grit SiC paper, 6 μm diamond paste, 0.3 μm alumina slurry and 0.05 μm alumina slurry. The resulting surface was then degreased with acetone and rinsed with distilled water. The coupons were then etched for 30 sec. with a reagent of 4 vol % concentrated HNO_3 and 10 vol % CH_3COOH diluted by H_2O.

The reference and counter electrodes used in these experiments consisted of 15 cm lengths of 0.8 cm dia. uranium rods. The molten salt electrolyte was the LiCl-KCl eutectic (44.2-55.8 wt %) with approximately 20 wt % UCl_3, which was generated <u>in situ</u> by the oxidation of metallic

uranium pins with $CdCl_2$. The $CdCl_2$ was reagent grade material from Cerac, Inc., Menomonee Falls, WI, and the anhydrous LiCl-KCl eutectic was from Lithcoa, Bessemer City, NC. Plating studies were conducted at 435°C.

Experimental Results

D.C. Plating Studies

The experimental conditions for these plating studies were varied over current densities of 5, 10, and 20 mA/cm^2 and plating times of 2.75 to 13.0 hours, producing uranium films between 100 and 330 mg/cm^2. During the course of these studies the coulombic efficiency of the plating process was found to be essentially 100 percent, and the quantities of uranium deposited are based on this observation. The results of these studies indicate that at higher current densities (20 mA/cm^2) there was a greater tendency to form dendrites, which were 2-3 mm in length. At lower current densities (5 mA/cm^2) and longer plating times, the electrodeposit was less dense with significant areas free of deposit. From these studies, it would appear that under constant-current plating conditions a 10 mA/cm^2 current density is near optimal. The uranium deposits formed at 10 mA/cm^2 had excellent bonding and may be adequate for fabrication of uranium targets for ^{99}Mo production.

Pulse-Plating Studies

A series of experiments were performed using the smaller Ni cathode to find the best pulse-plating conditions to provide the highest possible deposition rate for a dendrite-free uranium deposit. The condition was found to be pulsing at -65 mV for 0.1 msec followed by a 0.0 mV pulse for 1 msec. Using this regime, twenty-one hours are required to deposit a 0.3 mm (580 mg U/cm^2) film of uranium.

Under these conditions, a second series of experiments were performed to measure (1) the success of this method on electrodepositing uranium metal films on various substrates (stainless steel, carbon steel, zircaloy, nickel, and copper), and (2) benefits in uniformity gained by preanodization of the substrate and using a nucleating pulse. Nickel and copper substrates were clearly superior to the other metal coupons tested in bonding and uniformity. Overall, a nickel substrate was the superior of the two.

The use of preanodization of the nickel and a nucleation pulse both improve the uniformity of the deposit. The best deposit laid down to date is shown in Fig. 1. The deposit is nominally 0.3 mm thick, with a variation in thickness between 0 and 0.4 mm. This sample was prepared by use of a preanodization step of 5 coulombs at 1.35 V, followed by a -706 mA/cm^2, two-second nucleation pulse, followed by the 21-hour deposition. After deposition of uranium on the coupon in the helium atmosphere box, the solidified electrolyte was removed from the coupon by washing in the open atmosphere with 0.1N HCl, distilled water, absolute ethanol, and acetone.

The uranium appeared metallic with no signs of oxidation after two hours in the laboratory atmosphere. The bonding of the deposit to the substrate was excellent.

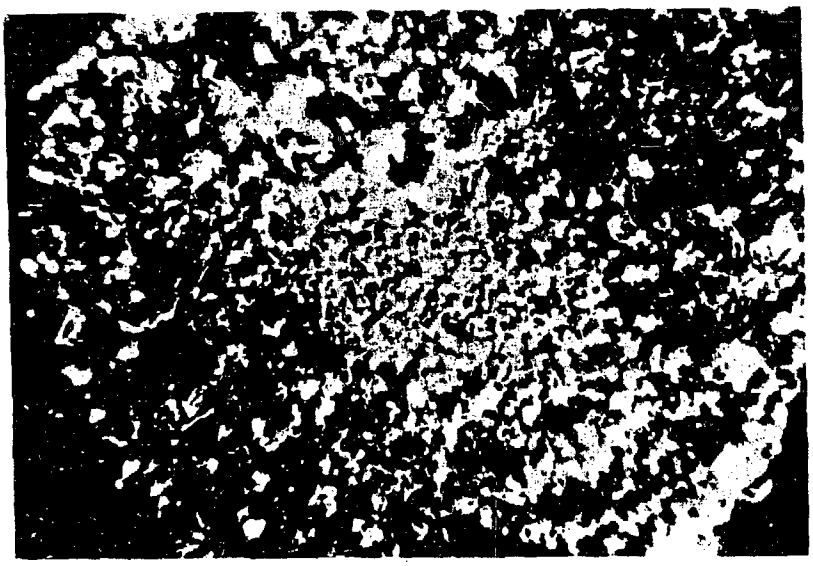

Fig. 1. The Best To-Date Uranium Deposit on Nickel Coupon

Further studies are needed to reduce our plating times and to improve the uniformity of the uranium deposit. In the latter case, studies are needed to optimize the nucleation pulse and preanodization regimes. Work is also needed to design and test the plating cell for the fabrication of the cylindrical targets. Studies are also needed to address nickel preplating of the stainless steel target cylinders. This nickel plating may well be done commercially by an electroless plating process.

Cost Analyses for Uranium Metal Target Fabrication

Cost analyses for the equipment and chemicals needed for an operational system completely set up for uranium-metal deposition on cylindrical target has been completed (Table 1). This system could electrodeposit uranium metal films on 2-6 cylinders simultaneously. Under our presently optimized plating regime, the system would produce 2-6 cylinders per day. Quotations on the major items (glovebox system, purification system, and furnace units) were received from Vacuum Atmospheres Company, Hawthorne, CA and from Lindberg, Watertown, WI, respectively. Quotations on the cost of nickel electroless preplating of the inner stainless steel target cylinders were received from TWR, Rosemont, IL. Cost estimates on the electrochemical plating equipment and the other smaller items, such as the plating cell and chemical cost, were derived from standard electrical units

Table 1. Equipment Cost for Target Fabrication

Item Description	Total Price
DXL-002-S-P Dri Lab Glovebox Stainless Steel, 90-in. wide x 30-in. deep x 33-in. high, with auxiliary equipment	$ 14,750
Mo-40-2V Dri-Train Dual Bed Purifier and Auxiliary Equipment for oxygen and moisture removal	16,815
Ni-40 Ni-Train and Auxiliary Equipment for nitrogen and/or hydrogen removal	19,460
Instrument A-1 Nitrogen Analyzer AM-2031 Moisture Analyzer AO-316-C Oxygen Analyzer AA-Audio Alarm ANL-1R Chart Recorder AV-1 Automatic Antechamber Control OP-30 Quick Purge	24,450
Electrical Connections, wiring, breakers, etc.	3,850
Two Reactor Furnace Tubes with connecting flange (6-in. O.D. x 30-in. length)	1,000
Fabrication and installation of two flanges in glovebox floor for furnace tubes	1,000
Factory Engineer for Start-up, demonstration and training of operators	2,850
Two Lindberg Model 54-M6024-Z Three Zone Hinged Furnace with controller and auxiliary equipment	16,554
Electrochemical Plating Equipment	10,000
Cell Housing, Chemicals, Ceramic Beakers etc.	15,000
TOTAL Cost of Target Fabrication System Facility	$125,729

and/or auxiliary equipment and chemicals and materials used here at the Argonne National Laboratory. The cost estimate from TWR on a 1 mil-thick nickel electroless preplating of the stainless steel or zircaloy target cylinders was $14 each.

In summary, it will cost approximately $120,000 to place into operation a uranium metal plating system for cylindrical targets for the production of ^{99}Mo. It should be noted that this cost includes certain items, such as the nitrogen and moisture analyzers, that are not really necessary but were included for completeness.

PROCESSING OF LEU METAL TARGETS

Patented processes for separating and purifying ^{99}Mo from cylindrical UO_2 film targets describe methods for dissolving the irradiated uranium material in acid and separating ^{99}Mo from uranium, transuranic elements, and other fission products.[1,5,6,25] The first separation/purification step for this process involves the use of either (1) precipitation of Mo by α-benzoinoxime or (2) adsorption of Mo by Ag-coated activated charcoal (ACAC) columns. Because the greater amount of uranium in LEU vs. HEU targets is especially important in this initial separation step, much of the work reported last year was directed to these two techniques. The results reported earlier showed that the precipitation process would not be hampered by the use of LEU, but the ACAC column separation was likely to be. This earlier work left two further concerns to be addressed. The first was the effect of higher amounts of uranium on the behavior of fission product ruthenium in these two separation techniques. The second was to attempt to optimize the less-than-satisfactory performance of the ACAC column separation for processing HEU or LEU targets. The following discusses our efforts in resolving these two concerns.

Separation of ^{99}Mo from Ruthenium

Experimental

The separation of ^{99}Mo from ruthenium was measured for both α-benzoinoxime precipitation and ACAC column adsorption under conditions similar to those presented elsewhere for processing of LEU and HEU targets.[1] Acidic solutions of natural molybdenum and depleted uranium were spiked with ^{103}Ru for these studies. Ruthenium-103 was measured by gamma counting using a GeLi detector.

Results

α-Benzoinoxime--The fraction of ^{103}Ru activity that remained with Mo after the precipitation of 0.5 mg Mo from 50 mL of solution by α-benzoinoxime was: 0.4% for 11.9 g U, 1.3% for 7.1 g U, and 3.5% for zero U. The trend in greater purification of Mo from Ru with greater amount of uranium indicates that use of LEU may reduce the chances for Ru contamination in the ^{99m}Tc product.

ACAC Columns--Four experiments were performed to test the effectiveness of ACAC column sorption of Mo for its separation from Ru. The results of this study, presented in Table 2, show that it is not as effective as the precipitation technique. Further, it appears that part of the Ru passes through the column during the Mo adsorption step, part is eluted with the Mo product, and part remains bound to the alumina after Mo is desorbed. There is no clear cut trend in the data, except that Ru is not very well separated from Mo by ACAC columns.

Table 2. Separation of Ru from Mo using ACAC Adsorption of Mo

Initial Conditions[a]		% Ru in			
[U],g	[H+],N	Mo Product	Eluted Feed	Remaining[b] on Column	Ratio % Ru Recovery[c] / % Mo Recovery
0	.6	10.9	53.9	35.2	0.13
7.12	0.6	7.2	45.3	47.5	0.13
7.12	0.4	5.6	42.0	52.4	0.075
11.90	0.4	10.9	38.0	51.1	0.19

[a]In 50 mL, 0.503 mg Mo in each experiment.
[b]Calculated from initial feed minus the sum of that in the eluted feed and the Mo product solution.
[c]In Mo product stream.

Optimization of ACAC Columns

Further studies were performed to attempt optimization of ACAC column recovery of Mo. Fresh column materials were prepared and separations were performed using instructions and suggestions in the literature.[5,6,25,26] As we reported last year, separations from uranium are not as good or as consistent as those attainable by the precipitation method, especially with uranium concentrations equivalent to those encountered by a first Mo separation step for processing of either LEU or HEU targets. Stripping of silver from the column material was noted for feeds containing high uranium concentrations. This stripping occurred with evolution of gas bubbles, which tend to destroy column effectiveness. As reported earlier, ACAC column separations are useful as a polishing step, after most of the uranium is removed by α-benzoinoxime precipitation of Mo. However, we conclude that these columns are not and will not be used commercially as a primary U/Mo separation and are planning no further work on them.

PROCESSING OF LEU SILICIDE TARGETS

It appears possible to convert current HEU targets that use aluminum alloy or uranium aluminide to LEU by use of uranium silicides without changing target geometries or sacrificing ^{99}Mo irradiation-yields. The following is a summary of our studies that have addressed modifications in current technology that may be necessary to process irradiated LEU silicide targets using basic dissolution. The current processing of HEU targets[27,28] is likely to include dissolving the cladding and meat of the irradiated target in 2-5M NaOH. In this step uranium and many of the fission products precipitate as hydroxides; ^{133}Xe is released and collected; and other species, including Mo, are solubilized in the aqueous solution. As the pH is decreased by the addition of acid, ^{131}I is released from solution as a gas. The acidified solution is then run through an alumina column, where Mo is sorbed. Molybdenum is subsequently eluted from the column and further purified.

Concerns in processing of LEU silicide targets stem from (1) the greater amount and different composition of uranium to be processed, (2) the presence of alloying elements that are necessary in clads for silicide fuel meats, and (3) the presence of significant quantities of silica in process solutions. The effects of these concerns on each processing step are discussed below.

Target Dissolution

Dissolution studies were performed using unirradiated miniplates and miniplate components (Al6061 cladding material, aluminum powder, and depleted uranium U_3Si and U_3Si_2). The miniplates and their components were supplied by R. F. Domagala and coworkers, Argonne National Laboratory.

Solid mixtures of Al6061 cladding, aluminum powder, and either U_3Si or U_3Si_2 were heated with a 3M NaOH solution. Dissolution of the cladding and aluminum powder was rapid but controlled; the uranium silicides appeared to not dissolve at all. The formation of copious amounts of black precipitate resulted from the precipitation of hydroxides of some of the alloying elements in Al6061 clad (nominal composition is reported in Table 3, all other elements are 0.05 wt % max with their total being 0.15 wt % max; the balance is Al). The major metallic constituent of the precipitate was aluminum (83 to 92 wt %). Other metals contained in the precipitate are also reported in Table 3. Likely due to some complexation by hydroxide ion, the precipitation of alloying element hydroxides was not complete; the extent of their precipitation is reported in the last column of Table 3.

More drastic measures than heating with concentrated base alone are necessary to dissolve the U_3Si and U_3Si_2 materials. A 1/1 3M NaOH/30% H_2O_2 solution readily dissolved these materials with heating. Hydrogen peroxide not only dissolves uranium; it also complexes it and prevents it from precipitating. After dissolution of the uranium silicides is complete, H_2O_2

Table 3. Fate of Alloying Elements in Al6061 Clad during Alkaline Dissolution of Uranium Silicide Targets

Element	Weight % of Composition			
	Al6061[a] Clad	Alkaline Precipitation[b]	Alkaline ppt, plus Supernant[b,c]	% of Element that Precipitated[c]
Al	95.9-98.6	83 - 92	96	28
Mg	0.8-1.2	4.6-8.0	1.1	99+
Si	0.4-0.8	2.9-4.5	1.8[d]	18
Fe	0.7 max	1.5-1.9	0.5	94
Cr	0.04-0.35	0.6-1.1	0.2	48
Cu	0.15-0.40	0.4-0.7	0.2	94
Mn	0.15 max	0.3-0.4	0.1	97+
Zn	0.25 max	0.2-0.3	0.1	67
Ti	0.15 max	0.1	0.04	78

[a]Based on manufacturing specifications, from R. Domagala, ANL.
[b]Range from two experiments, performed under slightly different conditions, dissolving both the requisite amount of Al6061 clad and Al powder for uranium-silicide dispersed fuel.
[c]Results from one experiment only.
[d]This higher than expected value is likely due to some dissolution of glassware.

must be destroyed to allow precipitation of uranium (thus separating it from the soluble molybdenum) and to prevent complications in downstream processing of the Mo. Destruction of the peroxide can be accomplished by the addition of hydroxylammonium nitrate (HAN), but its addition must be carefully controlled. If too much HAN is added, it too will complex uranium and prevent its precipitation. Addition of small quantities of acidic ferric nitrate solution (10^{-2} mol Fe/mol U) was found to catalyze the auto degradation of peroxide but does not take the reaction to completion. The addition of potassium permanganate will complete the destruction of H_2O_2 and allow complete precipitation of uranium hydroxide. Lowering the pH to ~8 will allow the precipitation of MnO_2.

If ^{99}Mo is not leached from the irradiated uranium silicide fuel during basic dissolution of the cladding and the aluminum powder dispersion material, it would be possible to have a two step process. Because of their much lower density, the precipitated alloying-element hydroxides can be easily swirled away, leaving the high density uranium silicides behind

for further processing. After the separation is complete, the silicides can be dissolved independently with alkaline peroxide. This process will be tested with low burnup targets in future experiments.

Acidification

Before molybdenum can be separated from the dissolver solution, the pH of the dissolver solution must be decreased with the addition of acid. Acidification to a pH of ~3 causes the precipitation of aluminum; this precipitate redissolves as more acid is added. To prevent the precipitation of gelatinous silica, the concentration of silicate in solution must be kept or lowered to ~0.1M.

Adsorption of Mo on Alumina

Molybdenum can be separated from other components of the acidified dissolver solution by its adsorption on a bed of alumina. Molybdenum can then be stripped from the column by a concentrated alkaline solution. Experiments were run to measure the effect on Mo adsorption by alumina of the concentrations of hydrogen ion, uranium, and silica. Representative data on their effects on the distribution ratio (Kd) between alumina and aqueous solutions are presented in Figs. 2, 3, and 4, respectively. The higher the distribution ratio, the better sorbed Mo is to the alumina and the smaller the alumina column must be. The sorption of Mo on alumina is extremely dependent on the concentration of Mo in solution. Molybdenum(VI) has an extremely complex solution chemistry due to polymer formation.[29,30] It appears that the MoO_4^{2-} monomer and smaller polymers sorb better than larger polymers. Therefore, Mo sorption is highest at lower Mo concentrations. Under conditions envisioned for processing LEU-silicide targets (essentially complete precipitation of uranium and [Si]$\geq$0.1M), the concentrations of these species will allow satisfactorily sorption of Mo on alumina columns.

Conclusions

Preliminary results lead us to conclude that a switch to LEU-silicide fuels will necessitate modifications in current processing schemes. These modifications do not appear overly dramatic, and it is likely that yield and purity of the ^{99}Mo will not vary significantly from current processing of HEU targets.

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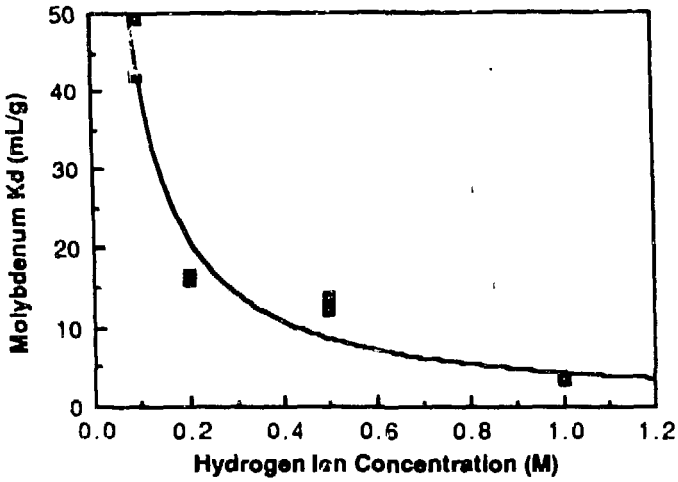

Fig. 2. Effect of $[H^+]$ on Mo Adsorption $[Mo] = 7 \times 10^{-4}$M.

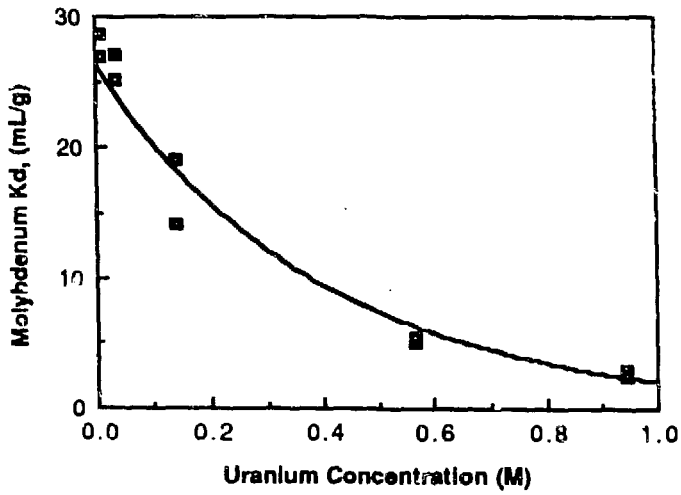

Fig. 3. Effect of Uranium Concentration on Mo Adsorption $[Mo] = 1.4 \times 10^{-3}$M, $[H^+] = 0.5$M.

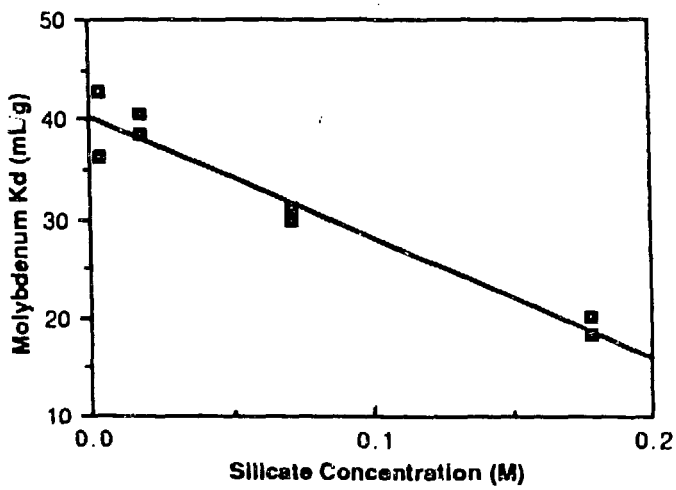

Fig. 4. Effect of Silicate Concentration on Mo Adsorption $[Mo] = 2.0 \times 10^{-3}$M, $[H^+] = 0.5$M.

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