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CRITICALITY ASPECTS OF THE
REVISED ZIRCONIUM DISSOLUTION SYSTEM
AT THE IDAHO CHEMICAL PROCESSING PLANT

W. G. Morrison

IDAHO NUCLEAR CORPORATION
NATIONAL REACTOR TESTING STATION
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The work was performed at the Idaho Chemical Processing Plant, operated by Phillips Petroleum Company under the auspices of the United States Atomic Energy Commission. A paper was presented at the Twelfth Annual Meeting of the American Nuclear Society in Denver, Colorado, June 20-23, 1966, which summarized the information contained in this report.

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CRITICALITY ASPECTS OF THE REVISED ZIRCONIUM
DISSOLUTION SYSTEM AT THE IDAHO CHEMICAL PROCESSING PLANT

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I. ABSTRACT

Soluble neutron poison has been used in a pioneering achievement at the Idaho Chemical Processing Plant as the primary nuclear safety control for routine processing of irradiated reactor fuel. Extensive calculations based on carefully controlled experiments have established concentration limits for the soluble boric acid poison. Special safety circuits and rigidly enforced operating procedures have been used to maintain nuclear safety at a level commensurate with the high standards used throughout the plant. This innovation has permitted safe use of large non-geometrically safe dissolution equipment for semi-continuous processing of enriched uranium-zirconium fuel with a resultant ten-fold increase in zirconium fuel processing capacity.

II. INTRODUCTION

The Idaho Chemical Processing Plant (ICPP) at the National Reactor Testing Station began using soluble neutron poison as the primary nuclear safety control in routine processing of irradiated, highly enriched, zirconium-uranium fuel elements late in 1964. Use of soluble poison permitted about a ten-fold increase in the zirconium fuel processing capacity. Fixed nuclear poisons have been used at ICPP for certain other applications for several years, but this is the first use of a soluble neutron poison.

Recovery of highly enriched uranium from zirconium-uranium alloy fuels is one of two principal processes employed at the Idaho Chemical Processing Plant, the other being processing of highly enriched aluminum-uranium alloy fuels. Aqueous processes with conventional solvent extraction equipment are used for processing these fuels, and over 99 percent of the uranium input is recovered. Soluble poison permits safe, efficient, and economical processing of several types of zirconium alloy fuel elements containing widely varying uranium loadings in a versatile facility with large, non-geometrically safe equipment. The zirconium fuels which are processed are typified by those from the Pressurized Water Reactor (PWR) at Shippingport, Pennsylvania. The gross uranium content may vary up to 3.0 weight percent, although most contain 1.5 to 2.0 percent uranium. To provide safe operation for all possible fuels, design calculations reported herein were based on fuel containing 3.3 weight percent uranium, 91 percent enriched, and no credit was taken for burnup or any burnable poison present.

III. DESCRIPTION OF ZIRCONIUM FUEL PROCESS

Processing facilities for zirconium fuels are installed in shielded cells at ICPP. The principal headend equipment for this process consists of a large diameter dissolver and large diameter tanks for continuous collection, composition adjustment, and storage of dissolver product solution--all adapted from the earlier batch zirconium process installation⁽¹⁾. The primary ICPP three-column solvent extraction system is used for first-cycle extraction operations. The first-cycle product is collected in intermediate storage and held for later processing through the remaining cycles of solvent extraction. A schematic diagram of the principal components of the dissolution and first-cycle extraction system and a chemical flowsheet of process conditions keyed to the equipment are presented in Figure 1.

The original zirconium fuel process used safe-mass limitation as the primary nuclear safety control for the dissolver. This limited the process capacity to about one kilogram of uranium per day. By use of a soluble poison in the dissolvent, and with a minor dissolver modification to permit semicontinuous fuel charging, the mass limitation could be removed and the potential capacity of the equipment more fully utilized. These changes yielded a capacity for the new process of about ten kilograms of uranium per day.

Boric acid was selected as the soluble poison compound for use in the process because of its low cost and because laboratory and pilot plant tests showed that its presence had no adverse effect on solution stability or other process variables. All process solutions are stable during normal operation; however, mal-operation could result in precipitation of solids. Calculated and experimental data presented herein indicate this poison to be capable of preventing criticality excursion in the dissolver charged full of fuel elements even with uranium accidentally precipitated out of solution during a dissolution. Operating experience has demonstrated that with normal plant control of dissolvent concentration and temperature, precipitate formation is no problem.

The reader is referred to other published reports for a broad description of the process concept and zirconium processing capability of the ICPP⁽²⁾ and for detailed record of the extensive development information for this dissolution process^(3,4,5).

1. Process Dissolver

The large-diameter dissolver, shown schematically in Figure 2, is operated in a semicontinuous manner. The fuel basket and charging chute of the dissolver are filled with fuel elements during an eight-hour charging cycle. During fuel charging, the dissolver is

STREAM	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17 & 18	19	20	
DESCRIPTION	FUEL CHARGE	DISSOLVENT	CHARGE WATER	DISSOLVER PRODUCT	COMPLEXING SOLUTION	COMPLEXED DISSOLVER PRODUCT	SCRUB RECYCLE 1 & R	1 & F	1 & X	1 & R	1 & P	1 & S	1 & B	1 & C	1 & C	1 & C	1 & X	1 & R	1 & P	FIRST CYCLE PRODUCT
FLOW, l/hr	517(a)		452(a)	27(a)	472(b)	60	530	500	530	500	60	500	200	500	200	10	200	2.1		
TBP, %								10.0		10.0		10.0		10.0						
DILUENT								Amsco		Amsco		Amsco		Amsco		Amsco				
Zr++++, M			1.07		0.80		0.71		0.71											
H ⁺ , M	6.54		6.35		4.76		4.22		4.22											
Al+++ , M				1.95	0.37	1.5	0.50		0.50		1.50									
H ⁺ , M	5.52		0.98	1.46	1.02		0.90		0.78	0.13	-1.13(d)		0.005		0.005		0.005	0.5		
NO ₃ , M	0.03		0.015	7.30	1.41	4.5	1.75		1.63	0.14	3.36	0.007	0.005		0.022		0.022	2.0		
Sn, g/l			1.53		1.14		1.01		1.01											
Cr, g/l				5.20	0.99		0.88		0.88											
B, g/l	3.80	7.0	3.84		2.90		2.58		2.58											
U, g/l			1.17(c)		0.88	0.201	0.78		<7.8x10 ⁻⁴	0.84		0.84			2.10		2.10	200		
DENSITY AT 25°C	1.035		1.186	1.360	1.215	1.230	1.220	0.794	1.200	0.802	1.200	0.795	0.998	0.794	1.002	0.757	1.002	1.275		
U, kg/batch	10(c)																			
Zr, kg/batch	835																			
Sn, kg/batch	13																			

(a) AVERAGE RATE FOR A 16-HOUR DISSOLUTION CYCLE.

(b) AVERAGE RATE FOR CONTINUOUS EXTRACTION (24-HOUR DAY).

(c) ASSUMES A 40% BURNUP OF URANIUM. (CRITICAL SAFETY ASSURED IN ABSENCE OF BURNUP, t_0 , FOR UP TO 16.7 K_g FULLY ENRICHED URANIUM.)

(d) THE USE OF DIBAN IN THE SCRUB MAKES IT 13M ACID DEFICIENT.

Figure 1: Schematic and Chemical Flowsheets of Zirconium Fuel Dissolution and First Cycle Extraction.

Figure 2. Zirconium Fuel Dissolver

filled with borated water to cushion the impact as elements fall into the dissolver. After fuel charging is complete, the charge water is removed and poisoned dissolvent--aqueous hydrofluoric acid containing dissolved boric acid as the nuclear poison--is fed continuously to the dissolver for 16 hours. Almost all of the fuel charge is dissolved during the dissolution cycle. A new charge of fuel is then added (on top of any fuel remaining from the previous charge) and the dissolving cycle is resumed. The primary modification made to the dissolver for semicontinuous operation was installation of a new fuel basket and charging chute.

The fuel basket, which has small slotted openings, is an extension of the fuel charging chute. Openings in the original fuel basket were too large to retain partially dissolved fuel elements. (Fuel retention in the basket was not required in the original process.) It was shown in pilot plant tests that an upright cylindrical dissolver with a solution annulus provided good circulation of dissolvent for efficient dissolution of fuel⁽⁵⁾. Also, this arrangement left no stagnant areas inside the dissolver for accumu-

lation of small pieces of undissolved fuel. Dissolver product solution flows continuously from the dissolver through an overflow line and a nitrogen gas lift to the downstream composition adjustment tank. The part of the fuel charge stacked up in the charging chute gradually drops, during the course of the dissolution cycle, into the dissolvent as the fuel at the bottom of the basket is dissolved. The ratio of dissolvent to metallic fuel in various portions of the dissolver is thus continually changing as the dissolution operation proceeds. Dissolver temperature control is maintained at 60-70°C by circulating borated cooling water through the dissolver jacket.

2. Composition Adjustment and Extraction

In the dissolver product composition adjustment step, aluminum nitrate is added continuously to the dissolver product in a large diameter tank which overflows alternately to either of two large diameter collection tanks for input measurements. Aluminum nitrate is added for the two-fold purpose of complexing free fluoride ions to prevent corrosion of the austenitic stainless steel (material of construction of all subsequent equipment) and of oxidizing the uranium to the extractable uranium-VI state. The first-cycle extraction system using the TBP-Amsco system operates continuously for separating uranium from the zirconium and the bulk of the fission products.

Soluble poison is required for nuclear safety only in the dissolver, since the uranium concentration in all downstream equipment is low--under two grams per liter. However, the presence of boron in the solution serves to guard against any hazard from the unlikely, but remotely possible, precipitation of uranium in the first-cycle extraction column. A combination of concentration control and safe geometry is used for primary nuclear safety control in the extraction operation.

3. Special Nuclear Safety Consideration in the Process

In addition to insuring nuclear safety during the dissolution step by addition of adequate boric acid to the dissolvent (covered in detail in the discussion of design calculations for various possible operating conditions), a number of other precautions were taken for nuclear safety reasons before this process was placed in routine operation.

3.1 Isolation from Plant

The dissolver is isolated from all sources of liquid that do not contain boron by sealing of lines to prevent accidental dilution of the poison. Beyond the dissolver, any inleakage

of water into the system would not present a nuclear hazard because of the low uranium concentrations; however, level alarms on the vessels would detect such a volume increase.

3.2 Cooling System

The dissolver, dissolver off-gas condenser, and acid feed tank are all cooled by a separate cooling system circulating borated water in a closed loop so that any leak in a heat transfer surface would not dilute the poison in the process solution. The borated coolant is circulated from a small surge tank through a heat exchanger. Plant cooling water or steam is used in the heat exchanger as the secondary heat transfer medium. Any loss or gain in the volume of borated coolant water in the closed system would be indicated by a level alarm on the surge tank. Any leakage from the dissolver to the borated water system would also be indicated by a radiation alarm on the circulating line.

3.3 Boron Monitor

Presence of nuclear poison in the dissolvent is assured by both monitoring instruments and by administrative control. Each batch of dissolvent prepared is analyzed in the laboratory prior to use in the dissolver. As a further check to guard against dilution, the boron concentration is continually monitored by two independently functioning neutron attenuation measurement instruments located in the dissolvent tank itself. If either instrument indicates less than the required boron concentration, a solenoid valve automatically closes to prevent any flow of dissolvent to the dissolver. The change in concentration is also signaled by an alarm.

3.4 Fuel Charging

Fuel cannot be charged directly into the dissolvent because of the safety problems which would be created by the release of hydrogen from the reaction. Also, it is not desirable to charge fuel directly into the empty dissolver because of mechanical shock limitations. Fuel is charged, therefore, into the dissolver filled with borated water to cushion the impact produced by the heavy fuel elements dropping about 20 feet. Charging water containing at least five grams of boron per liter is recycled between the dissolver and a hold tank. Fresh charging water is made up and added to the hold tank to compensate for heel loss and jet dilution of the boron concentration. Makeup charge water is analyzed during each charge cycle. Experience has shown that adjustment of the charge water to at least five grams of boron per liter gives complete confidence that the boron concentration when in the dissolver will be above the 3.8-gram-per-liter minimum requirement. (The boron requirement is established in Section IV, Design Calculations.)

4. Plant Operating Experience

Prior to processing irradiated fuel, 4962 kg of zirconium and unirradiated PWR fuel (uranium-235 alloyed with zirconium) were dissolved as a final proof-test of the process and equipment associated with the use of soluble poison. The operating procedures developed for control and use of the soluble poison were refined during this preliminary operation, and the procedures which evolved were used with only minor changes during subsequent operation with irradiated fuel.

Approximately 200 kg of uranium were recovered from irradiated PWR fuel elements in the first part of the processing campaign, with no problems developing attributable to the use of soluble poison. Routine processing rates of approximately 10 kg of uranium per day were sustained during normal operation. Special handling of a portion of this fuel was requested by the reactor operator for obtaining burnup data; this required a special dissolver cleanout after each dissolver batch, thus reducing the overall average processing rate to 7.5 kg of uranium per day. As mentioned earlier, dissolution rates for this fuel prior to the use of soluble poison were approximately one kilogram of uranium per day. Thus, the ten-fold increase in capacity predicted from development work was obtained in routine operation.

The use of this soluble poison dissolution process will be continued at ICPP in future processing of all enriched zirconium-uranium alloy fuels.

IV. DESIGN CALCULATIONS

1. Bases for Nuclear Design Calculations

Machine computations were used for all of the few-group multiregion design calculations for the zirconium process dissolver. All fuel loadings into the dissolver were considered as complete fuel elements composed of clad and meat rather than meat composition only. Laboratory studies showed that the zirconium-uranium alloy meat dissolves essentially as fast as it is exposed by dissolution of the cladding and that meat material will not accumulate. Also, during the dissolution cycle there is continuous movement of clad fuel assemblies from the charging chute to the dissolving zone.

Since the physical conditions inside the dissolver are not known precisely at any given time, the volume fraction concept was used for the design calculations. With this concept, complex mixtures in the basket inside the dissolver were considered as

simple homogeneous mixtures of an arbitrary metal volume fraction (MVF) and the complementary liquid volume fraction (LVF). The MVF giving maximum reactivity was determined, and the condition for maximum reactivity was used in all subsequent calculations. This conservative basis provides an extra margin of safety in the calculations for most of the expected operating conditions.

The uranium content of the dissolver product solution was assumed constant at 4.0 grams per liter for all of the calculations; this proved to be over twice the normal operating concentration and also provides additional conservatism in the calculations. Boron concentrations in the dissolver were varied in initial calculations to narrow the range for final consideration. The actual boron concentration in the dissolver varies according to the MVF present; however, all references to boron concentration refer to the boron concentration in the LVF. All curves in the figures for reactivity versus MVF or uranium concentrations are for constant boron concentrations in the LVF.

Nuclear constants for use in the few-group diffusion calculations were obtained according to the following tabulation:

<u>Computer</u>	<u>Codes</u>	
	<u>Fast Groups</u>	<u>Thermal Group</u>
IBM-650	MUFT-R ⁽⁶⁾	SOFOCATE ⁽⁷⁾
IBM-7040	GAM-1 ⁽⁸⁾	TEMPEST-40 ⁽⁹⁾

Nuclear constants obtained on the IBM-650 and used in the initial diffusion calculations for both the normal and abnormal conditions were for infinite systems. Nuclear constants used in subsequent diffusion calculations for plateout and reactivity of final design under normal operating conditions were for the finite system.

2. Scoping Calculations

Initial calculations were performed to determine the MVF in the central region or fuel basket for maximum reactivity of the dissolver. Dimensions were based on the dissolver as shown in Figure 2 without the 13-inch diameter fuel basket. These one-dimensional, two-group three-region calculations considered fuel mixture, annular solution, and unborated reflector water only. Nuclear constants used in these calculations are listed in Table I (Appendix). Figure 3 shows the results of these calculations in the upper curve for the major diameter section or most reactive part of the dissolver. Maximum reactivity with three grams of

Figure 3. k_{eff} Values for Normal Operation

normal boron per liter in the dissolvent and the basket full of fuel elements gave a k_{eff} of 0.91 which occurred at 0.6 MVF. A similar curve was obtained for the minor diameter section which showed the maximum reactivity also occurring at 0.6 MVF but with a slightly lower k_{eff} value. These two sections are actually stacked on top of each other. Instead of performing coupling calculations to determine the effect of one section on the other, the simpler approach of considering each section for infinite height was employed; calculated reactivities for infinite height were only two percent higher than those for the finite system height.

3. Nuclear Calculations for Normal Operating Conditions

Results of the scoping calculations indicated that the dissolver is critically safe for operation to produce dissolver product containing up to 4.0 grams U-235 per liter from zirconium-3.3 w/o uranium fuel, provided the metallic fuel remains in the fuel basket and that the boron concentration in the liquid phase is maintained at 3.0 grams per liter. Final design dimensions of the dissolver as shown in Figure 2 were then used for calculating the k_{eff} for normal operating conditions with 3.8 grams boron per liter in the dissolvent, a value that was selected for plant operation based on calculations for an abnormal operating condition discussed later. Diffusion codes as well as transport codes were used for these calculations. The following tabulation shows the results of these calculations at maximum reactivity condition in the fuel basket and including the effects of structural material and borated cooling water.

k_{eff} At Normal Operating Conditions by
Various Machine Codes for Maximum
Reactivity Conditions

<u>Code</u>	<u>Type</u>	<u>Computer</u>	<u>k_{eff}</u>
DMM(10)	Diffusion	IBM-650	0.797
DTK	Transport	IBM-7090	0.794
TOPIC(11)	Transport	IBM-7040	0.817
AIM-6(12)	Diffusion*	IBM-7040	0.805
FOG-3(13)	Diffusion	IBM-7040	0.789

*Used 16-group DTK constants

Results show very good agreement by the various calculational methods. The lower curve on Figure 3 shows the DMM results for an MVF range of 0.35 to 0.8. Nuclear constants used for the DMM and FOG-3 calculations are listed in Table II (Appendix).

Original specifications called for about twice the poison concentration in the coolant water as in the dissolvent. However, use of borated cooling water containing 8.0 grams boron per liter tended to give mechanical problems with pump seals. Recalculation of the dissolver with cooling water containing 5.0 grams boron per liter gave no significant change in the k_{eff} . Probably the only requirement is that the boron content in cooling water be at least 3.8 grams per liter to guard against diluting the boron in the dissolver in event of a leak into the system.

4. Nuclear Calculations for Abnormal Operating Conditions

Abnormal operating conditions had to be considered in the complete evaluation of this dissolver for nuclear safety. The boron concentration of 3.8 grams per liter used during plant operation was selected on the basis that even in the remote event of severe abnormal operating conditions, no nuclear incident could occur. Detailed investigation of four cases covered all nuclear safety situations during the worst conceivable abnormal operations. It is doubtful, however, that any of these situations would ever develop, much less be allowed to continue to the point that the assumed conditions were approached. The following tabulation lists the assumed abnormal conditions and the machine codes used in calculations for analyzing these conditions.

Calculations for Abnormal Operation Conditions

<u>Case</u>	<u>Computer</u>	<u>Code</u>
1. Precipitated $UF_4 \cdot 3/4 H_2O$ ^(a)	IBM-7094	PDQ-2(14)
2. Continued Dissolution while Precipitating $UF_4 \cdot 3/4 H_2O$ ^(b)	IBM-650	DMM
3. Undissolved Fuel in Annulus ^(b)	IBM-650	DMM
4. Plateout of $UF_4 \cdot 3/4 H_2O$ ^(b)	IBM-7040	FOG-3

(a) Cylindrical coordinates

(b) One dimensional

4.1 Precipitated $UF_4 \cdot 3/4 H_2O$ Settling Around Fuel Basket

The possibility of uranium precipitating presents the greatest problem and involves more complications than any of the other abnormal conditions considered. If an error is made in dissolver makeup resulting in a gross excess of fluoride ion in the dissolver, $UF_4 \cdot 3/4 H_2O$ will precipitate. The initial

investigation assumed that the error caused precipitation of all of the dissolved uranium present in the dissolver. Precipitated uranium was assumed to vary from completely dispersed to almost completely settled. A total of 16.1 kg U-235 was assumed in the dissolver; 13.0 kg as undissolved fuel in the basket and 3.1 kg precipitated. Undissolved fuel in the basket was assumed distributed at 0.6 MVF. Results of calculations for both 3.0 and 4.0 grams of boron per liter in the LVF are shown in Figure 4. Nuclear constants for these calculations are shown on Table III (Appendix) and the location of the compositions of Table III are shown on Figure 6 (Appendix). The reactivity appears to be essentially independent of the height of precipitate in the annulus. An interpolation showed that 3.8 grams of boron per liter were required to yield a k_{eff} of 0.9 for the precipitate settling around the basket. On the basis of this interpolation, a boron concentration of 3.8 grams per liter was selected for plant operation. This poison concentration was used in all subsequent calculations.

4.2 Continued Dissolution While Precipitating $UF_4 \cdot 3/4 H_2O$

An extension of the previous case considered additional precipitation of $UF_4 \cdot 3/4 H_2O$ in the event the error in dissolvent makeup was not detected during the dissolution cycle. Under such conditions, the zirconium-uranium alloy fuel would continue to dissolve and the dissolved uranium would precipitate continuously. As the fuel dissolves, its height in the basket decreases while the uranium concentration in the precipitate slurry builds up. For convenience in simulating maximum reactivity, the height of fuel in the basket and that of the precipitate slurry were assumed equal; this height was varied from 100 cm to 20 cm. Correspondingly, the U-235 concentration in the slurry varied from 26 to 240 grams per liter, while the boron concentration was held at 3.8 grams per liter in the LVF. The maximum k_{eff} calculated using nuclear constants listed in Table IV (Appendix) was 0.91 at 70 cm fuel height for about 50 grams U-235 per liter as precipitate in the liquid volume fraction. Results of the diffusion calculations for these conditions are shown in Figure 5. The reactivity was almost constant over the entire range of precipitate concentrations.

4.3 Undissolved Fuel in the Annulus

The third type of abnormal operating condition considered was that of a complete failure of the fuel basket to yield a uniform distribution of undissolved metallic fuel in the annulus. A calculation was made for this case which extended the 0.6 MVF mixture in the fuel basket over the entire cross-sectional area of the dissolver. Nuclear constants for this

Figure 4. k_{eff} Values for Concurrent Dissolution and Precipitation of $UF_4 \cdot 3/4 H_2O$

Figure 5. k_{eff} Values for Precipitation and Settling of $UF_4 \cdot 3/4 H_2O$

calculation are listed in Table V (Appendix). A resulting k_{eff} of 0.96 was obtained for a fuel height of 57 cm (equivalent to 17.6 kg U-235 in the dissolver) with 3.8 grams of boron per liter in the LVF. Vessel walls (Monel) and borated reflector water with 8.0 grams of boron per liter in the cooling water jacket were included in this calculation. An independent calculation for these same conditions yielded a k_{eff} of 0.95. Neglecting the Monel in the vessel walls resulted in a k_{eff} of 0.98. Considering that the assumed condition is very severe and several highly improbable events would be required to approach the assumption, these k_{eff} values are acceptable. First, the fuel basket would have to rupture for very much metallic fuel to get into the annulus. Second, the charge would have to exceed the normal charge appreciably. Finally, all of the fuel would have to distribute itself at 0.6 MVF. Practice has shown that the fuel charge stacks up the entire length of the charging chute instead of collecting near the bottom of the dissolver. In view of the unlikelihood of occurrence of these events, a calculated k_{eff} of 0.96 is reasonable.

4.4 Plateout of $UF_4 \cdot 3/4 H_2O$

Solid uranium fluoride material tended to coat the inside surfaces of the pilot plant dissolver under certain conditions⁽⁵⁾. An extensive series of calculations were therefore performed to evaluate the effect of possible plateout on the reactivity of the plant dissolver. Results indicate that an unusually large amount of uranium (90 to 100 kg) is necessary in order to present a hazard. An equal thickness of coating was assumed on each of the five internal surfaces available for plateout--two surfaces on the new basket, two on the original basket, and one on the dissolver vessel wall. Assumed conditions for these calculations were essentially the same as those used for previous abnormal operating conditions--the basket contained fuel mixture at 0.6 MVF, boron content of the LVF was 3.8 grams per liter, and vessel walls and borated reflector water were included. A separate region adjacent to each surface in the dissolver was included for plateout material. The dimension search option in the FOG code was used to determine the thickness of the plated-out material necessary for a specific k_{eff} value. A sample of plateout material from the pilot plant work was used as the basis for physical and nuclear properties of the plateout regions in the calculations. In addition, the literature values for physical properties of $UF_4 \cdot 3/4 H_2O$ (which differed slightly from the pilot plant material) were used in a similar calculation as a comparison. Results showed very little difference in mass of uranium for the same reactivity; however, there was some difference in thickness of the plated-out material for the same reactivity. It was assumed in both cases that no boron was occluded in the plateout. Four-group

nuclear constants for all of these calculations are shown in Table VI (Appendix). Assumed plateout containing 50 kg of uranium with an additional 22 kg of uranium in the fuel basket at 0.6 MVF yielded a calculated k_{eff} of 0.91 with 3.8 grams of boron per liter in the dissolvent.

5. Correlation of Experimental Data with Computational Methods

Clean critical experiments performed at Oak Ridge National Laboratory Critical Facility applicable to the poisoned dissolver system⁽¹⁵⁾ were used to check the calculational methods used for the dissolver design. A detailed comparison⁽¹⁶⁾ of various calculational methods for correlating the experimental results showed that the calculations agree very well with the experimental values. Results for the experiments with fuel element arrays with dissolved boric acid and uranyl nitrate agreed within 0.5 percent. Calculated k_{eff} values for some arrays with buildup six-foot-long elements (three fuel element tiers high) neglecting the 1-1/8-inch gaps between fuel in the plates were all higher than the experimental values. In general, this close agreement between calculated and experimental values gives complete confidence in the nuclear design calculations for the use of soluble poison as the primary nuclear safety control in the zirconium dissolver system.

V. PLANT TESTS

1. Prestartup Neutron Multiplication Measurements

Several complete unirradiated PWR fuel subassemblies and many metallurgical specimens containing in total about 33 kg U-235 were available for nonradioactive tests. A portion of this material was used for in-situ neutron multiplication measurements as part of the startup tests. These measurements were made with the complete subassemblies and with other subassemblies fabricated from whole fuel plates included among the metallurgical specimens. The average uranium content of these subassemblies was 2.0 percent, appreciably under the 3.3 percent used in all design calculations.

Three fuel element arrays were assembled containing progressively more uranium up to 9.9 kg U-235. The arrays were lowered into the empty dissolver, and water containing 3.8 grams of boron per liter was added in measured increments. Count rates were obtained after each incremental addition of the borated water. Also, a count rate meter was observed during the addition so that the addition could be stopped immediately if necessary. If all factors employed in the dissolver design calculations could have been duplicated in the test,

a multiplication of three to five was expected for the largest array. The actual multiplication measured, however, was less than two. The most probable reasons for not measuring the calculated multiplication were the physical inability to position the fuel over the entire 13-inch diameter basket region and to duplicate the uranium concentration used in the calculations. The borated water did not contain dissolved uranyl nitrate, and only about 70 percent of the basket region cross-sectional area was occupied because of the geometrical impossibility of filling a circular basket with these particular square fuel elements. The MVF occupied by complete PWR fuel elements was close to 0.6, which yields maximum reactivity, but the uranium concentration was appreciably less than the 3.3 percent used in the calculations.

2. Cold Dissolution Measurements

Instrumentation for the neutron multiplication measurements was left in place in order to monitor the dissolver during actual operation. It was believed that this instrument could monitor any unexpected buildup of an undissolved heel or buildup of insoluble $\text{UF}_4 \cdot 3/4 \text{H}_2\text{O}$. Background count rates taken during the dissolver checkout dissolutions with zirconium metal showed no indication of change because of fuel shifting around during dissolution. Count rates measured during dissolution of the unirradiated PWR fuel were also nearly constant; however, there were slight changes which were similar in all runs when correlated with percent of dissolution time. However, the magnitude of the changes was not believed indicative of any uranium buildup. From all indications, the equipment functioned perfectly. Complete dissolver cleanouts at the completion of a significant period of PWR fuel processing produced less than 2.9 kg of uranium, so it is not surprising that no significant multiplication was measured during the dissolution operations.

VI. CONCLUSIONS

Soluble boron has been successfully used as a neutron poison for primary criticality control in large scale processing of irradiated nuclear reactor fuel without any unusual problems. The low capacity of a batch process was increased about ten-fold by use of the soluble neutron poison and semicontinuous operation.

Diffusion calculations were shown to be adequate for nuclear design of chemical processing equipment. Confidence in such calculations was provided by the excellent agreement with measured results from clean critical experiments performed at ORNL.

Use of carefully prepared procedures and proper instruments to guarantee the presence of the soluble poison in the process streams has been an essential part of this effective control of nuclear safety.

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VIII. APPENDIX

Tables I - VI present the few group averaged macroscopic cross sections used in the diffusion calculations for the criticality safety design of the zirconium dissolver. Figure 6 shows the location of the compositions listed in Table III.

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Table I
NUCLEAR CONSTANTS FOR SCOPING CALCULATIONS

Composition		One Fast Group	Thermal
0.2 MVF - 0.8 LVF	D	1.2538	0.1720
3.0 g boron/liter in LVF	Σ_a	0.00415	0.1796
	Σ_r	0.03752	--
	$\nu\Sigma_f$	0.002337	0.1286
0.35 MVF - 0.65 LVF	D	1.2534	0.1988
3.0 g boron/liter in LVF	Σ_a	0.0050	0.2051
	Σ_r	0.03017	--
	$\nu\Sigma_f$	0.00404	0.2250
0.5 MVF - 0.5 LVF	D	1.2537	0.2353
3.0 g boron/liter in LVF	Σ_a	0.005825	0.2306
	Σ_r	0.02268	--
	$\nu\Sigma_f$	0.005676	0.3214
0.6 MVF - 0.4 LVF	D	1.2524	0.2682
3.0 g boron/liter in LVF	Σ_a	0.00634	0.2476
	Σ_r	0.01761	--
	$\nu\Sigma_f$	0.006699	0.3857
0.8 MVF - 0.2 LVF	D	1.2450	0.3723
3.0 g boron/liter in LVF	Σ_a	0.006930	0.2816
	Σ_r	0.007421	--
	$\nu\Sigma_f$	0.008253	0.5143
Dissolver Product	D	1.2589	0.1480
(Annular Solution 0.020 MVF)	Σ_a	0.002832	0.1515
3.0 g boron/liter	Σ_r	0.04644	--
	$\nu\Sigma_f$	0.000236	0.1286
Coolant Water	D	1.2558	0.1530
	Σ_a	0.000803	0.01966
	Σ_r	0.04893	--

Table II
NUCLEAR CONSTANTS FOR NORMAL OPERATING CONDITIONS
(DMM Calculations)

Composition		1 Fast Group	Thermal
0.35 MVF - 0.65 LVF	D	1.2651	0.1962
3.8 g boron/liter in LVF	Σ_a	0.005075	0.2273
	Σ_r	0.02771	--
	$\nu\Sigma_f$	0.003848	0.2255
0.5 MVF - 0.5 LVF	D	1.2740	0.2323
3.8 g boron/liter in LVF	Σ_a	0.005708	0.2478
	Σ_r	0.02013	--
	$\nu\Sigma_f$	0.005302	0.3222
0.6 MVF - 0.4 LVF	D	1.2815	0.2649
3.8 g boron/liter in LVF	Σ_a	0.006003	0.2615
	Σ_r	0.01510	--
	$\nu\Sigma_f$	0.006128	0.3866
0.7 MVF - 0.3 LVF	D	1.2912	0.3082
3.8 g boron/liter in LVF	Σ_a	0.006159	0.2752
	Σ_r	0.01007	--
	$\nu\Sigma_f$	0.006725	0.4510
0.8 MVF - 0.2 LVF	D	1.2983	0.3685
3.8 g boron/liter in LVF	Σ_a	0.005985	0.2889
	Σ_r	0.005298	--
	$\nu\Sigma_f$	0.006836	0.5155
Dissolver Product	D	1.2428	0.1435
(Annular Solution 0.020 MVF)	Σ_a	0.003286	0.1855
3.0 g boron/liter	Σ_r	0.04617	--
	$\nu\Sigma_f$	0.0002322	0.01204
Monel	D	0.5909	0.2378
	Σ_a	0.002755	0.3520
	Σ_r	0.0001046	--
Borated Coolant Water	D	1.2509	0.1348
(4.8 w/o H_3BO_3)	Σ_a	0.005812	0.3551
	Σ_r	0.04486	--

Table II (Con't)
NUCLEAR CONSTANTS FOR NORMAL OPERATING CONDITIONS
(FOG-3 Calculations)

Region	Composition		Three Fast Groups			Thermal
			Group 1	Group 2	Group 3	
1	0.6 MVF - 0.4 LVF 3.8 g boron/liter in LVF	D	1.8672	1.0276	0.7837	0.4084
		Σ_a	0.00074	0.00096	0.01990	0.1602
		Σ_r	0.06282	0.05992	0.04852	--
		$\nu\Sigma_f$	0.001093	0.001418	0.01986	0.2338
2, 4 6, 8	Monel	D	1.3780	0.7900	0.2702	0.2326
		Σ_a	0.00049	0.001455	0.01404	0.2175
		Σ_r	0.02642	0.00241	0.00019	--
3, 5	Dissolver Product (Annular Solution 0.02 MVF) 3.8 g boron/liter	D	2.0633	1.0637	0.6096	0.1903
		Σ_a	0.00061	0.00020	0.00727	0.1115
		Σ_r	0.1041	0.1442	0.1429	--
		$\nu\Sigma_f$	0.00004	0.00005	0.00071	0.007793
7	Borated Coolant (4.8 w/o H_3BO_3 Solution)	D	1.8788	1.0364	0.6010	0.1757
		Σ_a	0.000586	0.000299	0.01367	0.2320
		Σ_r	0.1073	0.1446	0.1388	--

Table III
 NUCLEAR CONSTANTS FOR DISSOLVER CALCULATIONS
 PRECIPITATED $UF_{4.3/4}H_2O$ SETTLING AROUND
 14 5/8-INCH DIAMETER FUEL BASKET (IBM-7090 PDQ-RZ)

Composition		1 Fast Group	Thermal
1. 0.6 MVF - 0.4 LVF 3.0 g boron/liter in LVF	D	1.2524	0.2682
	Σa	0.00634	0.2476
	Σr	0.01761	--
	$v\Sigma f$	0.006699	0.3857
1. 0.6 MVF - 0.4 LVF 4.0 g boron/liter in LVF	D	1.2527	0.2652
	Σa	0.00657	0.2644
	Σr	0.01742	--
	$v\Sigma f$	0.006682	0.3857
2. Supernate Solution 3.0 g boron/liter	D	1.2547	0.1458
	Σa	0.002708	0.1456
	Σr	0.04652	--
2. Supernate Solution 4.0 g boron/liter	D	1.2560	0.1436
	Σa	0.003326	0.1876
	Σr	0.04598	--
3. 0.6 MVF - 0.4 Ppt. Solution 220.4 g U-235/liter 3.0 g boron/liter in LVF (Ppt. height = 5.0 cm)	D	1.3020	0.250
	Σa	0.00765	0.384
	Σr	0.01180	--
	$v\Sigma f$	0.00950	0.680
3. 0.6 MVF - 0.4 Ppt. Solution 170.3 g U-235/liter 3.0 g boron/liter in LVF (Ppt. height = 10.0 cm)	D	1.2995	0.2576
	Σa	0.007017	0.3246
	Σr	0.01307	--
	$v\Sigma f$	0.008052	0.5463
3. 0.6 MVF - 0.4 Ppt Solution 137.0 g U-235/ liter 3.0 g boron/liter in LVF (Ppt height = 30.0 cm)	D	1.2906	0.2677
	Σa	0.006438	0.2732
	Σr	0.01460	--
	$v\Sigma f$	0.006954	0.4393
3. 0.6 MVF - 0.4 Ppt Solution 130.3 g U-235/liter 3.0 g boron/liter in LVF (Ppt height = 50.0 cm)	D	1.2853	0.2698
	Σa	0.006305	0.2629
	Σr	0.01516	--
	$v\Sigma f$	0.006771	0.4179
3. 0.6 MVF - 0.4 Ppt Solution 137.0 g U-235/liter 4.0 g boron/liter in LVF (Ppt height = 30.0 cm)	D	1.2811	0.2646
	Σa	0.006807	0.2900
	Σr	0.01513	--
	$v\Sigma f$	0.007139	0.4393
4. Precipitate Solution 250.4 g U-235/liter 3.0 g boron/liter (Height 5.0 cm)	D	1.1220	1.282
	Σa	0.0125	0.460
	Σr	0.0388	--
	$v\Sigma f$	0.01285	0.830
4. Precipitate Solution 125.2 g U-235/liter 3.0 g boron/liter (Height 10 cm)	D	1.1268	0.1363
	Σa	0.007126	0.3390
	Σr	0.04111	--
	$v\Sigma f$	0.006749	0.4016

Table III (Con't)

4. Precipitate Solution	D	1.1305	0.1435
41.7 g U-235/liter	Σ_a	0.004349	0.2103
3.0 g boron/liter	Σ_r	0.04380	--
(Height 30 cm)	$v\Sigma_f$	0.002318	0.1339
4. Precipitate Solution	D	1.1348	0.1450
25.0 g U-235/liter	Σ_a	0.003938	0.1845
3.0 g boron/liter	Σ_r	0.04440	--
(Height 50 cm)	$v\Sigma_f$	0.001445	0.08031
4. Precipitate Solution	D	1.1307	0.1413
41.7 g U-235/liter	Σ_a	0.005001	0.2522
4.0 g boron/liter	Σ_r	0.04380	--
(Height 30 cm)	$v\Sigma_f$	0.002390	0.1389
5. Monel	D	0.5909	0.2378
	Σ_a	0.002755	0.3520
	Σ_r	0.000105	--
6. Borated Coolant Water	D	1.2509	0.1348
4.8 w/o H_3BO_3	Σ_a	.005812	0.3551
	Σ_r	.04486	--
7. Pure Water	D	1.2558	0.1530
	Σ_a	0.000803	0.01966
	Σ_r	0.04893	--

Figure 6

CYLINDRICAL DISSOLVER COMPOSITION LOCATION
FOR ALL CASES

Table IV
 NUCLEAR CONSTANTS FOR DISSOLVER CALCULATIONS
 CONTINUED DISSOLUTION WHILE PRECIPITATING $UF_4 \cdot 3/4 H_2O$
 (Six-Region DMM on the IBM-650)

Region	Composition		1 Fast Group	Thermal
1	0.6 MVF - 0.4 Ppt. Solution	D	1.2792	0.267
	130.8 g U-235/liter	Σ_a	0.00655	0.278
	3.8 g boron/liter in LVF	Σ_r	0.01537	--
	Precipitate height = 100 cm	$v\Sigma_f$	0.00689	0.420
2,4,6	Monel	D	0.5909	0.2378
		Σ_a	0.00275	0.3520
		Σ_r	0.000105	--
3	Precipitate Solution	D	1.272	0.1463
	26.3 g U-235/liter	Σ_a	0.00414	0.2160
	3.8 g boron/liter	Σ_r	0.0436	--
	Precipitate height = 100 cm	$v\Sigma_f$	0.00131	0.0830
5	Borated Coolant Water 4.8 w/o H_2BO_3 (8 g boron/liter)	D	1.2509	0.1348
		Σ_a	0.005812	0.3551
		Σ_r	0.04486	--
1	0.6 MVF - 0.4 Ppt. Solution	D	1.2806	0.264
	139.8 g U-235/liter	Σ_a	0.00677	0.292
	3.8 g boron/liter in LVF	Σ_r	0.01502	--
	Precipitate Height = 70 cm	$v\Sigma_f$	0.00724	0.459
3	Precipitate Solution	D	1.276	0.1439
	49.0 g U-235/liter	Σ_a	0.00487	0.252
	3.8 g boron/liter	Σ_r	0.0429	--
	Precipitate height = 70 cm	$v\Sigma_f$	0.00265	0.155
1	0.6 MVF - 0.4 Ppt. Solution	D	1.2826	0.2607
	151.9 g U-235/liter	Σ_a	0.00704	0.3098
	3.8 g boron/liter in LVF	Σ_r	0.01455	--
	Precipitate height = 50 cm	$v\Sigma_f$	0.00770	0.4875
3	Precipitate Solution	D	1.2791	0.1408
	79.1 g U-235/liter	Σ_a	0.00583	0.3008
	3.8 g boron/liter	Σ_r	0.04215	--
	Precipitate height = 50 cm	$v\Sigma_f$	0.004382	0.2539
1	0.6 MVF - 0.4 Ppt. Solution	D	1.2872	0.2526
	179.9 g U-235/liter	Σ_a	0.00763	0.3331
	3.8 g boron/liter in LVF	Σ_r	0.01359	--
	Precipitate height = 30 cm	$v\Sigma_f$	0.008725	0.5775
3	Precipitate Solution	D	1.2707	0.1350
	149.2 g U-235/liter	Σ_a	0.00794	0.4090
	3.8 g boron/liter	Σ_r	0.04037	--
	Precipitate height = 30 cm	$v\Sigma_f$	0.007931	0.4789
1	0.6 MVF - 0.4 Ppt. Solution	D	1.2269	0.2431
	215.0 g U-235/liter	Σ_a	0.00820	0.4072
	3.8 g boron/liter in LVF	Σ_r	0.01251	--
	Precipitate height = 20 cm	$v\Sigma_f$	0.00986	0.6900
3	Precipitate Solution	D	1.1865	0.1284
	236.8 g U-235/liter	Σ_a	0.01026	0.5442
	3.8 g boron/liter	Σ_r	0.03826	--
	Precipitate height = 20 cm	$v\Sigma_f$	0.01201	0.7602

Table V
NUCLEAR CONSTANTS FOR DISSOLVER
CALCULATIONS WITH UNDISSOLVED FUEL IN ANNULUS
(Six-Region DMM Problem on the IBM-650)

Region	Composition		1 Fast Group	Thermal
1,3	0.6 MVF - 0.4 LVF 3.8 g boron/liter in LVF	D	1.2526	0.2661
		Σ_a	.006525	0.2611
		Σ_r	.01746	--
		$\nu\Sigma_f$	.00669	0.3857
2,4,6	Monel	D	0.5909	0.2378
		Σ_a	0.002755	0.3520
		Σ_r	0.000104	--
5	Borated Coolant Water 4.8 w/o H_3BO_3	D	1.2509	0.1348
		Σ_a	0.005812	0.3551
		Σ_r	0.04486	--

Table VI
NUCLEAR CONSTANTS FOR DISSOLVER CALCULATIONS
WITH PLATED-OUT $UF_4 \cdot 3/4 H_2O$ ON INTERNAL SURFACES
(13 Region FOG-3 on the IBM-7040)

Region	Composition		Three Fast Groups			Thermal
			Group 1	Group 2	Group 3	
1	0.6 MVF - 0.4 LVF 3.8 g boron/liter in LVF	D	1.86718	1.02757	0.78370	0.4084
		Σ_a	0.0007395	0.0009602	0.019902	0.1602
		Σ_r	0.062818	0.059922	0.048516	--
		$\nu\Sigma_f$	0.001093	0.0014178	0.019865	0.2338
2,4 6,8 10	Plate-out $UF_4 \cdot 3/4 H_2O$ $\rho = 4.242 \text{ g/cm}^3$ (Pilot Plant Basis)	D	2.75433	1.35521	0.94772	0.1034
		Σ_a	0.01015	0.01542	0.12548	2.734
		Σ_r	0.03874	0.02382	0.001382	--
		$\nu\Sigma_f$	0.02705	0.03331	0.19774	5.625
3,7 11,13	Monel	D	1.3780	0.7900	0.2702	0.2326
		Σ_a	0.00049	0.001455	0.01404	0.2175
		Σ_r	0.02642	0.00241	0.00019	--
5,9	Dissolver Product (Annular Solution 0.020 MVF) 3.8 g boron/liter	D	1.89303	0.97855	0.59999	0.1903
		Σ_a	0.000737	0.000129	0.007873	0.1115
		Σ_r	0.10728	0.15189	0.14112	--
		$\nu\Sigma_f$	0.000038	0.000047	0.000711	0.007793
12	Borated Coolant Water (4.8 w/o H_3BO_3)	D	1.87885	1.03640	0.60105	0.1757
		Σ_a	0.000586	0.000299	0.01367	0.2320
		Σ_r	0.1073	0.1446	0.1388	--
12	Borated Coolant Water (3.0 w/o H_3BO_3)	D	1.8858	1.04114	0.6041	0.1442
		Σ_a	0.000574	0.000185	0.008887	0.1913
		Σ_r	0.10726	0.14474	0.1422	--
2,4 6,8 10	Plate-out $UF_4 \cdot 3/4 H_2O$ $\rho = 5.98 \text{ g/cm}^3$ (Literature value AERE-C/R-1346)	D	1.95212	0.96230	0.67191	0.07333
		Σ_a	0.01430	0.02171	0.17561	3.854
		Σ_r	0.05461	0.03357	0.001951	--
		$\nu\Sigma_f$	0.03813	0.04696	0.2788	7.930
Fission Source		X	0.75356	0.24644	0.0	0.0