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SOLUBLE NEUTRON POISONS AS A PRIMARY CRITICALITY CONTROL
IN SHIELDED AND CONTAINED RADIOCHEMICAL FACILITIES

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ABSTRACT

Studies indicated that the use of soluble poisons as a primary criticality control offers economic and other advantages in that it permits the factors of vessel size and shape and solution concentrations to be dictated by considerations other than those of criticality. It is believed that soluble poison criticality control can be made as reliable as other methods of conditional control if the application is preceded by adequate development work and is monitored by multiple, independent safeguards.

The studies included multigroup machine calculations of the required content of poisons in solutions of fissile and fertile material, a compilation of data on the detection, stability, decontamination, and costs of soluble poisons, and an assessment of the possible effects of a nuclear excursion.

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1.0 INTRODUCTION

This report presents the results of studies on the use of soluble neutron poisons for primary criticality control in shielded, remotely operated radiochemical facilities. The concentration of poisons required in solutions of fissile materials, instrumentative and procedural safeguards for assuring the required poison concentrations, stability of soluble poisons in solution, removal of poisons from fissile products, economic considerations in the use of soluble poisons, and the effects of accidental nuclear excursions are discussed.

In the evolution of nuclear fuel processing it has been recognized that the use of nuclearly geometrically safe equipment, i.e., vessels of area to volume ratio such that neutron leakage prevents a sustained nuclear reaction, imposes a large penalty in cost and capacity to chemical processing plants, while experience has shown that the increased expenditure per unit of capacity may not be justified in light of the relatively minimal effects of an accidental nuclear excursion in a remote, shielded area.

This cognition led to the establishment of a practical rule, now used to some extent in all fuel processing facilities, which stated that at least two independent human or mechanical operating errors must be made before a nuclear accident can occur. This rule, in effect, allowed a finite probability of a nuclear accident but provided that this probability have a suitably low value. The rule permitted the use of batch or concentration control provided that two or more independent methods of ensuring the batch size or concentration were in effect.

The rule has sometimes been interpreted to mean that neutron poisons in solutions of fissile material could not be used as a primary criticality control since one error, i.e., forgetting to add the poison, would result in a nuclear excursion. It is possible, however, that even in the use of soluble neutron poisons the probability of an accidental excursion may be made as low as required. This low probability can be achieved by providing that two or more unrelated errors must be made before poisons are left out of a solution or before a physical or chemical process for removing the poison from solution can occur.

The use of soluble poisons is particularly applicable in moderate- or large-scale standard chemical processing equipment that is difficult or impractical to modify for inclusion of fixed poisons and is severely limited by the requirements of concentration or batch control. It is ordinarily possible to include sufficient poison that an infinite volume of the solution could not sustain a nuclear reaction; thus, one is not concerned with neutron interaction or the use of geometry-safe catch basins, which are typical of geometry-safe or fixed-poison-safe installations.

Considerations of the use of soluble poisons as a primary criticality control have been published previously in ORNL correspondence and reports (1-4). These considerations led to an ORNL managerial endorsement (5) of the use of soluble neutron poisons as a primary control in a remotely operated, shielded, contained facility in which due precautions were to be taken to ensure the addition of poison and the stability of the poison in solution.

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2.0 SUMMARY

Studies of the use of soluble poisons for criticality control included multigroup machine calculations of the poison content required for solutions of fissile and fertile material in natural water, a compilation of data on the detection, stability, decontamination, and costs of soluble poisons, and an assessment of the possible effects of a nuclear excursion in a well-shielded, well-contained radiochemical facility. Results indicated that soluble poison criticality control is feasible and offers advantages in that the factors of vessel size and shape and solution concentrations may be dictated by considerations other than those of criticality. One or more of the possible neutron poisons probably will be stable at the required concentration in most process solutions and can be adequately removed from the fissile product.

Required Poison Contents. Multigroup machine searches for the concentrations of B, Cd, Sm, and Gd required to cause $k_{\infty} = 1$ for natural water solutions of U^{235} , U^{233} , Pu^{239} , 5% U^{235} - U^{238} , and 10% U^{235} -Th were made. The calculations indicated that the atomic ratio of the high-thermal-cross-section poisons to the fissile materials for $k_{\infty} = 1$ continually increases with concentration because of the harder neutron spectrum and greater dependence on the relatively lower cross sections for high-energy neutrons. In a natural water solution of hydrogen to U^{235} atom ratio of 50 (~500 g of U^{235} per liter), 1.02, 0.23, 0.0144, and 0.035 atoms of B, Cd, Sm^{149} , and Gd, respectively, per atom of U^{235} are calculated to cause $k_{\infty} = 1$.

The calculations indicated that U^{233} and Pu^{239} are surprisingly more reactive than U^{235} at high concentrations. Ten or more times as much poison per atom of fissile material was required for U^{233} and Pu^{239} as for U^{235} in concentrated solutions.

Safeguard Techniques. Safeguard techniques for providing a high degree of assurance of the safe soluble poison concentration in solutions may be of the procedural and/or instrumentative types. The procedural type of safeguard would consist of independent batch additions of poisons, perhaps backed up by an independent chemical analysis. A neutron absorptiometer appears to be a very satisfactory instrument for in-line measurement of poison concentrations.

Stability. The limits of stability of soluble poisons in most solutions of interest have been defined roughly, but it is recommended that tests be performed with actual process solutions before the poisons are used for primary criticality control. Boron is soluble to the extent of 0.2-0.5 M in most solutions. Cadmium, samarium, and gadolinium are generally soluble to concentrations of 1-3 M in nitrate solutions.

Loss of soluble poisons by evaporation does not appear to be a problem in typical aqueous solution processes.

Poisons are expected to be deposited and included in corrosion films to an extent less than 1% of the weight of the film.

Removal of Soluble Poisons from Solutions. Experimental data indicated that the soluble poisons of interest may be satisfactorily decontaminated from fissile and fertile materials in typical fuel

processing solvent extraction procedures.

Costs. Soluble poison control may significantly decrease plant and equipment costs since it may use indiscriminate vessel shape and greater throughput for a given vessel size. The material cost of the poison is generally negligible, of the order of < \$1 per kilogram of fissile material.

Effects of a Nuclear Excursion. A water-moderated nuclear excursion in a radiochemical facility that meets ORNL criteria for shielding, containment, and ventilation is not expected to cause significant personnel exposures or levels of downwind ground contamination. The most serious effect of the maximum credible nuclear explosion would be the in-cell contamination that could result from rupture of a process vessel.

3.0 REQUIRED CONCENTRATION OF SOLUBLE POISONS

The concentrations of B, Cd, Sm, and Gd required to cause an infinite multiplication factor of 1 in natural-water solutions of U^{235} , U^{233} , Pu^{239} , 5% enriched uranium, and 10% U^{235} -90% Th were calculated (Figs. 1-5).

3.1 Calculational Procedure

All the calculations were made by the IBM-7090 MODRIC neutron diffusion code and a 34-group cross-section structure as used by Nestor (6). Cross sections for infinite dilution and 20°C, recently prepared by Nestor, were used for U^{235} , U^{233} , Pu^{239} , natural boron, cadmium and Sm^{149} . His resonance energy cross sections for U^{238} and Th^{232} were multiplied by the ratio of the effective resonance integral for a given scattering cross section per unit of absorber to the infinitely dilute resonance integral to correct for dilution. A machine program to obtain the Nestor group structure from available GNU cross sections for Gd was written.

The MODRIC concentration search routine was used to determine the poison concentration required to cause $k_{\infty} = 1.0000$ with fixed concentrations of fissile and fertile material in water. Three mesh points and a condition setting the space derivative of the flux at the boundary at zero were used. An average of 21 seconds of 7090 time was required to calculate each poison concentration with the initial poison concentration estimates based on a one-group calculation.

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Fig. 1. Concentration of boron required cause $k_{\infty} = 1.0000$ for solutions of boron and fissile and fertile materials in natural water.

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Fig. 2. Concentration of cadmium required to cause $k_{\infty} = 1.0000$ for solutions of cadmium and fissile and fertile materials in natural water.

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Fig. 3. Concentration of Sm^{149} required to cause $k_{\infty} = 1.0000$ for solutions of Sm^{149} and fissile and fertile materials in natural water (isotopic concentration of Sm^{149} in naturally occurring Sm is 13.84 wt %).

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Fig. 4. Concentration of natural Gd required to cause $k_{\infty} = 1.0000$ for solutions of Gd and fissile and fertile materials in natural water.

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Fig. 5. Effect of natural water solution concentration on the weight of natural B, Cd, Gd, and Sm required per unit weight of U^{235} to cause $k_{\infty} = 1.0000$.

3.2 Errors and Safety Factors

In general, the accuracy of the required poison content is expected to decrease with the H/X ratio because of neutron spectrum hardening and greater dependence on the fast cross sections. The most accurate calculations are believed to be those with pure U²³⁵, for which the cross sections are relatively well known, and boron, which shows 1/v behavior. It is believed that the accuracy of the B-U²³⁵ relation decreases from ~5% at low concentrations (H/U²³⁵ = 1000-2000) to perhaps 10-20% at high concentrations (H/U²³⁵ $\approx$ 30).

The calculated concentrations of Cd, Sm, and Gd, which depart radically from 1/v behavior, are likely to be underestimates of the concentration required for $k_{\infty} = 1$, especially at high concentrations, because of the lack of information for making a dilution correction. This effect is probably compensated to some degree by the vagaries of the neutron diffusion model at the high concentrations.

Extrapolations to zero poison concentrations indicate limiting critical concentrations in water of ~12.3, 11.8, and 9.1 g/liter for pure U²³⁵, U²³³, and Pu²³⁹, respectively. The experimentally determined current best estimates of these values are 12.1 (7), 11.2 (7), and ~8.5 (8), respectively.

In choosing a poison concentration for a practical application, it is recommended that the vessel size be considered infinite and that the curves in Figs. 1-4 be displaced toward higher H/X values by a factor of at least 1.5, consistent with use of at least 100% excess poison. The minimum recommended "safe" curve for U²³⁵ and boron is shown in Fig. 6 as an example. With this method of determining the safety factor, addition of boron to solutions of U²³⁵ would begin at concentrations of 8.1 g of U²³⁵ per liter. Until the required poison contents can be verified by possible future experiments, it is recommended that concentrations not be allowed to closely approach the present calculations modified by the recommended safety factors without benefit of neutron multiplication measurements.

The effect of neutron leakage in finite vessels that contain solutions with infinite multiplication factors of 1 was also determined with the MODRIC code. The effect of vessel size on the effective multiplication

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Fig. 6. Comparison of present calculations (solid line) of boron content required to cause $k_{\infty} = 1$ for solutions of enriched uranium with experimental values from HW-66882 (dashed line).

Fig. 7. Effect of diameter on the effective multiplication factor, k_{eff} , of bare spheres containing natural water solutions of U^{235} and B for which $H/U^{235} = 400$ and $k_{\infty} = 1.0000$.

factor of a solution of H/U²³⁵ ratio equal to 400 of sufficient boron content to cause a k_{∞} of 1 is illustrated in Fig. 7. The effect of solution concentration on the effective multiplication factor in a finite vessel is shown in Fig. 8.

Fig. 8. Effect of solution concentration on the effective multiplication factor, k_{eff} , in a 5-ft-dia bare sphere filled with natural water solutions of U²³⁵ and B for which $k_{\infty} = 1.0000$.

These data tend to indicate that the effective multiplication factor in vessels greater than a few feet in diameter, which are typical of those desired for use in fuel processing plants, is not significantly different from the infinite system multiplication factor. In general, the neutron leakage effect will not be sufficient to compensate for k_{∞} calculational errors of perhaps 50-100%.

3.3 Comparison with Experiments

A comparison of the present calculations with the only known directly applicable experiments (9) is also shown in Fig. 6. The experimental measurements of the boron content required for $k_{\infty} = 1$ were determined with 3% enriched UO_3 in polyethylene and 3% enriched $UO_2(NO_3)_2$ solutions in the Physical Constants Testing Reactor. The present values calculated with pure 3% enriched U, B, and H_2O , which should be comparable to the UO_3 -polyethylene- B_4C experiments, show ~20% more boron required than indicated by the experiments at high concentrations ($H/U^{235} = 400$ or $H/U \approx 12$) and tend toward progressively better agreement at lower concentrations.

These discrepancies are not unexpected for calculations on low-enrichment uranium because of the inadequacy of the multigroup cross sections in simulating the many important resonances in U^{238} . Better agreement is expected between calculations and experiments for pure U^{235} , U^{233} , Pu, and 10% U^{235} -90% Th^{232} .

The effect of the nitrogen in decreasing the reactivity in the experiments was significant. This effect is indicative of the lowered reactivity that will occur in many plant situations due to ionic species, such as those of nitrogen or chlorine, which have moderate cross sections.

Calculations of the effective multiplication factor of experimentally determined just-critical spheres containing U^{235} , U^{233} , and Pu^{239} solutions were performed as an aid in determining the reliability of the MODRIC calculational procedure and cross sections (Table 1). The calculated multiplication factors are in good agreement with the multiplication factors of 1.00 that were observed experimentally. The deviations of the calculated multiplication factors from those experimentally observed result from errors in the neutron leakage calculation and the infeasibility of duplicating all the conditions of the experiment as well as possible

Table 1. Calculated Values of k_{eff} for Experimentally Determined Just-critical Spheres Containing Solutions of U^{235} , U^{233} , and Pu^{239} in Natural Water

Reference to experiment	(10)	(11)	(11)	(12)	(10)	(12)
Sphere volume, liters	174	16.98	17.02	11.2	174	34.1
Sphere radius, cm	34.6	15.95	15.95	13.88	34.6	20.12
Elemental solution concentrations, atoms/barn·cm						
U^{233}	4.355×10^{-5}	1.73×10^{-4}	1.008×10^{-4}	4.291×10^{-4}	-	-
U^{234}	7.3×10^{-7}	9.3×10^{-7}	5.4×10^{-7}	-	-	-
U^{235}	2.6×10^{-7}	6.9×10^{-8}	4×10^{-8}	-	4.864×10^{-5}	-
U^{238}	2.9×10^{-7}	1.24×10^{-6}	7.2×10^{-7}	-	3.56×10^{-6}	-
Th^{232}	2.0×10^{-7}	-	-	-	-	-
F	-	3.46×10^{-4}	2.02×10^{-4}	8.582×10^{-4}	-	-
Al	4.8×10^{-6}	1.93×10^{-4}	1.93×10^{-4}	-	-	-
N	1.17×10^{-4}	-	-	-	1.77×10^{-4}	-
Fe	-	4.10×10^{-5}	4.10×10^{-5}	-	-	-
H	$6.6/4 \times 10^{-4}$	6.591×10^{-2}	6.683×10^{-2}	6.565×10^{-2}	6.707×10^{-2}	6.693×10^{-2}
O	$3.33/ \times 10^{-2}$	3.330×10^{-2}	3.361×10^{-2}	3.368×10^{-2}	3.353×10^{-2}	3.346×10^{-2}
Pu^{239}	-	-	-	-	-	6.630×10^{-5}
Al clad thickness, cm	0.32	-	-	-	0.32	-
H_2O reflector thickness, cm	0	0	7.62	0	0	0
Atoms H/atom fissile element	1532	381	663	153	1379	980
Experimental k_{eff}	1.0003	1.000	1.000	1.000	1.0001	1.00
Calculated k_{eff}	0.986	0.976	0.973	0.990	0.978	0.899

errors in cross sections. Of these sources of error, cross section errors are the only kind that would adversely affect the results of the present k_{∞} calculations.

4.0 PROCEDURAL AND INSTRUMENTATIVE SAFEGUARDS FOR ENSURING SOLUBLE POISON CONTENT

Multiple means must be provided for ensuring the soluble poison content of solutions in which the soluble poison is the primary criticality control. The concentration of poison in solutions that are subsequently to contain fissile material may be ensured by carefully prepared and recorded batch additions of poisons to known volumes of solution, chemical analysis by a variety of techniques, and by on-stream instrumentation. On-stream instrumentation and off-stream chemical analysis may also be used to routinely verify the poison content of process solutions.

The use of these safeguards must be re-inforced by knowledge of the stability of the required poison concentration under all normal and possible abnormal process conditions. This will ordinarily permit the use of process equipment in such a manner that the inadvertent separation of poison from fissile material by such mechanisms as excessive evaporation, deposition, or addition of incompatible chemicals will have a suitably low probability of occurrence.

4.1 Procedural Safeguards

A typical procedure for ensuring poison concentration in a solution that will subsequently contain fissile material is to provide for two independent additions of poison, either of which is sufficient to render the solution "safe." Each independent addition must be followed by a solution volume measurement and a suitable mixing treatment. This procedure is similar to the "Batch Card" method now used for concentration and mass control in many plants that process fissile material without shielding.

The use of independent batch additions may be supplemented by an independent off-stream analysis of poison concentration. Many analytical techniques are available for reliable analysis of neutron poisoning elements (13,14). The macroscopic neutron-absorption cross sections of

solutions of all such poisons can be measured by neutron absorptiometry.

4.2 On-stream Instrumentation

Development of on-stream analyzers for neutron poison concentration has been centered around the use of neutron absorptiometry techniques, since the technique is relatively simple to apply and furnishes a direct measurement of the macroscopic neutron-absorption cross section. Many installations, including ORNL (15), Hanford Laboratories (16,17), Argonne National Laboratory (18), and Callery Chemical Company (19), have developed and tested an on-stream neutron absorptiometry instrument, and many other installations, including the Union Carbide ORGDP Plant (16) and the Savannah River Plant (20-23), have developed related instruments.

The neutron absorption instruments that have been developed may be used to continuously monitor the neutron-absorption cross section of a flowing stream by means of a capsule neutron source (usually a Pu, Po, or Ra-Be source) on one side of the stream and a neutron detector (usually a BF_3 counter) on the other side of the stream. The instrument, which may use continuous standardization, can be made very reliable and has adequate sensitivity. The ANL instrument is capable of measuring a boron concentration of 0 to 2.65 g/liter with a maximum absolute error of 0.05 g/liter. The instrument can be made relatively insensitive to temperature. In high gamma radiation fields an instrument using a BF_3 detector can discriminate between gamma and neutron radiation if the gamma radiation is selectively shielded with lead to levels of 10-250 rad/hr.

5.0 STABILITY OF SOLUBLE POISONS IN SOLUTION

The stability of solutions of soluble poisons is dependent on the limits of solubility and the effects of evaporation and deposition by corrosion, all of which may be affected by radiation-induced chemical changes.

5.1 Solubility of Neutron Poisons

Solubility data for neutron poisons in typical aqueous fuel processing solutions are given in Table 2. Boron as H_3BO_3 or borax is generally soluble to the extent of 0.2-0.6 M in sulfuric or nitric acid

solutions at room temperature, and cadmium and rare earths are generally more soluble than boron.

Relatively little information is available on the solubility of nuclear poisons in organic liquids. A literature survey (30) showed that a boron chelate with a 1,2-dihydroxy compound, a compound of the type $(RBO)_n$, or the cadmium salt of a strong organic acid might be applicable for "poisoning" organic solutions.

5.2 Effects of Evaporation

Laboratory studies at ORNL have demonstrated that B, Cd, and rare earths are not volatilized significantly in Thorex or Darex evaporation-feed adjustment operations. Table 3 lists the data of Moore and Rainey (31) on the combined volatilization and entrainment of boron in the evaporation of simulated dissolver solutions in laboratory glassware. It is seen that the boron loss to the distillate is low even in the case where the evaporation was allowed to proceed to an abnormally high temperature of 181°C . The losses of cadmium and rare earths in similar experiments were less than 0.5%.

Goode (25) demonstrated that boron, added to 5 M HNO_3 -2 M HCl Darex dissolvent as 0.06 M boric acid, remained with the uranium during normal operation of the dissolution and chloride removal steps as well as during two cases of maloperation in which the still was allowed to evaporate to dryness.

5.3 Deposition in Corrosion Products

The deposition of boron and cadmium from reactor cooling water onto reactor core materials has been measured at Westinghouse (32) and Chalk River (14). Both studies indicated that when dilute water solutions of the poisons are exposed to metals, a few milligrams of boron or cadmium is included in each gram of metal corrosion product. This factor is important in nuclear reactors due to the decreased reactivity that accompanies poison inclusion on fuel elements but will probably cause insignificant loss of boron from process solutions in fuel processing facilities.

The Chalk River work hypothesized that the mechanism of the observed deposition of boron and cadmium on the corrosion product films of Zircaloy, stainless steel, and aluminum is ion exchange. The boron and

Table 2. Solubility of Neutron Poisons in Typical Fuel Processing Solutions

Solvent	Solute	Temp., °C	Solubility of B, Cd, or E.E., <u>g</u>	Ref.
4 <u>M</u> H ₂ SO ₄	H ₃ BO ₃	25	0.38	2
3.73 <u>M</u> H ₂ SO ₄	3 CdSO ₄ ·8H ₂ O	25	0.87	2
2.95 <u>M</u> H ₂ SO ₄	Na ₂ B ₄ O ₆ ·10H ₂ O	25	0.50	2
4 <u>M</u> H ₂ SO ₄ + 5 g stainless steel/liter	H ₃ BO ₃	25	0.36	2
3.15 <u>M</u> H ₂ SO ₄ + 42 g stainless steel/liter	H ₃ BO ₃	25	0.56	2
3.0 <u>M</u> H ₂ SO ₄ + 42 g stainless steel/liter	3 CdSO ₄ ·8H ₂ O	25	0.87	2
1.80 <u>M</u> H ₂ SO ₄ + 42 g stainless steel/liter	Na ₂ B ₄ O ₇ ·10H ₂ O	25	0.78	2
6 <u>M</u> H ₂ SO ₄	H ₃ BO ₃	25	0.27	2
5.55 <u>M</u> H ₂ SO ₄	3 CdSO ₄ ·8H ₂ O	25	0.24	2
5.0 <u>M</u> H ₂ SO ₄	Na ₂ B ₄ O ₇ ·10H ₂ O	25	0.40	2
6.0 <u>M</u> H ₂ SO ₄ + 5 g stainless steel/liter	H ₃ BO ₃	25	0.27	2
5.4 <u>M</u> H ₂ SO ₄ + 24 g stainless steel/liter	H ₃ BO ₃	25	0.64	2
4.88 <u>M</u> H ₂ SO ₄ + 24 g stainless steel/liter	3 CdSO ₄ ·8H ₂ O	25	0.78	2
3.70 <u>M</u> H ₂ SO ₄ g stainless steel/liter	Na ₂ B ₄ O ₇ ·10H ₂ O	25	0.76	2
93 wt % H ₂ SO ₄	H ₃ BO ₃	25-90	> 0.1	2
13 <u>M</u> HNO ₃	H ₃ BO ₃	25	0.33	2
60% HNO ₃	H ₃ BO ₃	20	0.215	24

Table 2, continued

Solvent	Solute	Temp., °C	Solubility of B, Cd, or R.E., <u>M</u>	Ref.
60% HNO ₃	H ₃ BO ₃	25	0.333	24
60% HNO ₃	H ₃ BO ₃	30	0.384	24
60% HNO ₃	H ₃ BO ₃	40	0.624	24
13 <u>M</u> HNO ₃ -0.04 <u>M</u> F ⁻ -0.04 <u>M</u> Al ³⁺	H ₃ BO ₃	25	0.22	2
10.6 <u>M</u> HNO ₃ -0.04 <u>M</u> F ⁻ -0.04 <u>M</u> Al ³⁺	Cd(NO ₃) ₂ ·4H ₂ O	25	1.34	2
8 <u>M</u> HNO ₃ -1 <u>M</u> Th ⁴⁺ -0.06 <u>M</u> U(VI)	H ₃ BO ₃	25	0.2	2
8 <u>M</u> HNO ₃ -0.9 <u>M</u> Th ⁴⁺ -0.05 <u>M</u> U(VI)	H ₃ BO ₃	25	0.22	2
8 <u>M</u> HNO ₃ -0.9 <u>M</u> Th ⁴⁺ -0.05 <u>M</u> U(VI)	Na ₂ B ₄ O ₇ ·H ₂ O	25	0.26	2
8 <u>M</u> HNO ₃ -0.9 <u>M</u> Th ⁴⁺ -0.05 <u>M</u> U(VI)	Cd(NO ₃) ₂ ·4H ₂ O	25	1.22	2
13 <u>M</u> HNO ₃ -0.04 <u>M</u> F ⁻ -0.1 <u>M</u> Al ³⁺	H ₃ BO ₃	23	0.26	2
13 <u>M</u> HNO ₃ -0.05 <u>M</u> F ⁻ -0.1 <u>M</u> Al ³⁺	H ₃ BO ₃	40	0.46	2
13 <u>M</u> HNO ₃ -0.04 <u>M</u> F ⁻ -0.1 <u>M</u> Al ³⁺	H ₃ BO ₃	60	0.71	2
13 <u>M</u> HNO ₃ -0.04 <u>M</u> F ⁻ -0.1 <u>M</u> Al ³⁺	H ₃ BO ₃	85	1.18	2
50% NaOH	H ₃ BO ₃	20	2.47	24

Table 2, continued

Solvent	Solute	Temp., °C	Solubility of B, Cd, or R.E., <u>M</u>	Ref.
50% NaOH	H ₃ BO ₃	30	2.61	24
50% NaOH	H ₃ BO ₃	40	2.63	24
H ₂ O	CaSO ₄	20	3.65	2
H ₂ O	CaSO ₄	60	4.0	2
H ₂ O	CaSO ₄	100	2.9	2
H ₂ O	H ₃ BO ₃	20	0.8	2
H ₂ O	H ₃ BO ₃	60	2.4	2
H ₂ O	H ₃ BO ₃	100	6.5	2
43% NaNO ₃	H ₃ BO ₃	20	0.76	24
43% NaNO ₃	H ₃ BO ₃	30	1.10	24
43% NaNO ₃	H ₃ BO ₃	40	1.37	24
5 M HNO ₃ -2 M HCL -.05 M NaF	H ₃ BO ₃	25-95	> 0.06	25
1 M HNO ₃ -0.126 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	1	> 0.37	26
1 M HNO ₃ -0.126 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	20	0.74	26
1 M HNO ₃ -0.252 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	1	> 0.37	26
1 M HNO ₃ -0.252 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	20	0.74	26
1 M HNO ₃ -0.504 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	1	> 0.37	26

Table 2, continued

Solvent	Solute	Temp., °C	Solubility of B, Cd, or R.E., M	Ref.
1 M HNO ₃ -0.504 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	20	0.74	26
0.126 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	1	> 0.37	26
0.252 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	1	> 0.37	26
0.252 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	20	0.74	26
0.504 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	1	> 0.37	26
0.504 M UO ₂ (NO ₃) ₂	H ₃ BO ₃	22	0.74	26
0.88 M Th-0.032 M U -0.51 M Al-5.7 M HNO ₃	CdF ₂	22	> 0.016	27
Above with 0.002 M PO ₄	CdF ₂	22	> 0.016	27
Above with 0.012 M PO ₄	CdF ₂	22	> 0.016	27
Above with 0.023 M PO ₄	CdF ₂	22	> 0.016	27
Above with 0.230 M PO ₄	CdF ₂	22	> 0.016	27
1.58 M Th-0.047 M U -0.80 M Al-(0.6 M HNO ₃)	CdF ₂	22	> 0.032	27
Above with 0.002 M PO ₄	CdF ₂	22	> 0.032	27
Above with 0.012 M PO ₄	CdF ₂	22	> 0.032	27
Above with 0.023 M PO ₄	CdF ₂	22	> 0.032	27
Above with 0.230 M PO ₄	CdF ₂	22	> 0.032	27
H ₂ O	Y(NO ₃) ₃	~25	3.8	28
1 N HNO ₃	Y(NO ₃) ₃	~25	3.6	28
2 N HNO ₃	Y(NO ₃) ₃	~25	3.3	28
3.8 N HNO ₃	Y(NO ₃) ₃	~25	2.8	28

Table 2, continued

Solvent	Solute	Temp., °C	Solubility of B, Cd, or R.E., <u>M</u>	Ref.
6.9 <u>N</u> HNO ₃	Y(NO ₃) ₃	~25	2.3	28
10.8 <u>N</u> HNO ₃	Y(NO ₃) ₃	~25	1.3	28
14.2 <u>N</u> HNO ₃	Y(NO ₃) ₃	~25	0.70	28
H ₂ O	Dy(NO ₃) ₃	~25	3.2	28
H ₂ O	Sm(NO ₃) ₃	~25	3.2	28
14.2 <u>N</u> HNO ₃	Sm(NO ₃) ₃	~25	0.47	28
H ₂ O	Gd(NO ₃) ₃	~25	1.6	28
14.2 <u>N</u> HNO ₃	Gd(NO ₃) ₃	~25	0.30	28
H ₂ O	Gd ₂ (SO ₄) ₃	0	0.131	29
H ₂ O	Gd ₂ (SO ₄) ₃	34	0.075	29
H ₂ O	Sm ₂ (SO ₄) ₃ ·8H ₂ O	25	0.028	29
H ₂ O	SmCl ₃	10	3.0	29
H ₂ O	SmCl ₃	50	3.4	29
H ₂ O	SmF ₃ ·1/2H ₂ O	~20	Insoluble	29
H ₂ O	Sm(OH) ₃	~20	Insoluble	29
H ₂ O	Sm ₂ O ₃	~20	Insoluble	29
H ₂ O	Sm ₂ (C ₂ O ₄) ₃ ·10H ₂ O	~20	Insoluble	29
H ₂ O	Gd ₂ O ₃	~20	Insoluble	29
H ₂ O	NH ₄ BF ₄	16	2.1	29
H ₂ O	NH ₄ B ₅ O ₈ ·4H ₂ O	18	0.24	29
H ₂ O	CdF ₂	25	0.29	29

Table 3. Volatility of Boron During Evaporation of Dissolver Solutions

Run No.	Initial Solution Composition, M						Init. Vol., ml	Vol. After Distillation % of Orig.	Max. Aq. Temp., °C	Boron, % by Analysis	
	B	HNO ₃	U	Th	Al(NO ₃) ₃	NaF				In Residue	In Distillate
1	0.1	-	-	-	-	-	500	5	101	95	< 1
2	0.1	8.5	-	-	-	-	500	5	120.5	96	3.6
3	0.2 ^a	8.2	0.06	0.95	-	-	500	51	119.5	> 99	< 1
4	0.04	8.2	0.06	0.89	0.04	0.04	520	37	135	97	3
5	0.2	8.2	0.06	0.95	0.04	0.04	500	25	138	93	2
6	0.05	4.4	0.02	0.5	0.04	0.02	2000	13	142	98	2
7	0.03	8	0.06	1	0.04	-	500	18	181	94	6
8	1.5	9.4	0.06	1.0	0.1	0.04	210	29	138	b	2

^aBoron added as Na₂B₄O₇; all other runs used H₃BO₃.

^bBoric acid crystallized on cooling.

cadmium content of corrosion product films exposed to solutions of cadmium sulfate and boric acid in heavy water at B and Cd solution concentrations of the order of 0.01 g/liter was independent of solution concentration and time of contact. The extent of boron and cadmium inclusion was 2-6 and 20-30 mg per gram of corrosion product, respectively, for stainless steel, Zircaloy, and aluminum. These data should apply, at least for an initial approximation, with higher concentrations of soluble poisons that may be used for criticality control.

The Westinghouse studies indicated that boron deposition on stainless steel from boric acid solutions at $\sim 310^{\circ}\text{C}$ containing approximately 1 g of B per liter of solution is of the order of 3 mg per gram of stable corrosion product film. The deposition appeared to be a factor of ~ 3 lower at 25°C .

5.4 Radiation Effects

Radiation levels up to a few watts per liter probably will have negligible direct effect on the stability of typical process solutions that may contain soluble poisons. A secondary effect may be due to increased deposition rates caused by increased corrosion rates that may accompany radiation-induced peroxide formation.

It has been shown at Bettis (33), Argonne (34), and Chalk River (14) that solutions of borates and boric acid tend to have greater radiolytic decomposition rates than pure water. The extent of this effect in fuel processing solutions that contain high concentrations of many ions is not known.

6.0 REMOVAL OF SOLUBLE POISONS FROM SOLUTIONS

Ion exchange and solvent extraction techniques for selective removal of soluble poisons from solutions have been investigated. An ion exchange process was developed for removing cadmium sulfate from the CANDU reactor cooling water (14). Moore and Rainey (31) studied the behavior of B, Cd, and Sm-Gd in typical fuel processing solvent extraction flowsheets.

In batch extraction tests Moore and Rainey showed the distribution coefficients of boron, cadmium, and Sm-Gd between aqueous nitrate solutions and 2.5% TBP in Amsco to be suitably low ($DC \text{ o/a} < 0.001$) under most test conditions for separation of the poisons from uranium. Batch countercurrent

demonstrations of the Interim-23 and TBP-25 flowsheets indicated that two extraction cycles should always be sufficient to decrease the nuclear poison content of a uranium product to an acceptable level.

7.0 COSTS OF SOLUBLE POISON CONTROL

Savings in capital plant and equipment investment can be realized through the use of soluble poison criticality control since the shape, size, spacing, and location of vessels can be dictated by process and operational requirements rather than by those of criticality. The operational costs in soluble poison control would be higher than in the use of strict geometry control but perhaps not significantly higher than in the use of mass or concentration conditional controls. All conditional control systems, which are believed to be the only systems feasible in future moderate- to large-scale plants, require accurate volume measurement and multiple determinations of solution composition.

As an example of savings that could be realized in equipment costs, recent estimates (35) of the cost of a stainless steel tank for storing solutions of U^{233} and Pu^{239} indicated that the cost of a standard tank suitable for soluble poison use was \$725 as compared to \$3900 for a geometrically safe pipe system. The costs of addition of fixed poisons to the tank would increase the tank cost to \$1400 for Pyrex glass raschig ring fixed poison and to \$3800 for a boron-stainless steel network of 1/8-in.-thick plates on 1.5-in. centers with a boron-stainless steel cost of \$3 per pound. These cost differentials become increasingly significant when spread over the many vessels in a fuel processing plant.

The cost of instrumentation in a moderate-to large-scale soluble-poison-controlled plant is probably not significantly higher than in a mass or concentration-controlled plant. The soluble-poison plant would be characterized by a relatively few vessels possessing neutron absorptiometers and liquid level, density, and sampling instruments as contrasted with a multitude of similarly equipped vessels, without neutron absorptiometers, in mass- or concentration-controlled plants.

The actual cost of the soluble poison, generally < 1\$ per kilogram of fissionable material, is ordinarily negligible compared with other operational costs, which are now of the order of \$1000-3000 per kilogram

of fissionable material for fuels of enrichment greater than a few per cent. The current cost of granulated technical grades of boric acid, borax, cadmium nitrate, gadolinium oxide, and samarium oxide in 100-lb lots is \$0.0878 (36), \$0.0531 (36), \$0.91 (36), \$44 (37), and \$33 (37) per pound, respectively. These costs and the data in Fig. 5 were used to construct Fig. 9, which compares the relative costs of the poison per kilogram of U^{235} required to cause $k_{eff} = 1$ as a function of solution concentration. The cost of B, Cd, Gd, and Sm per unit weight of U^{235} varies approximately as 1:10:25:100.

When criticality is not of primary consideration, it is usually desirable to operate fuel processing plants at fissile plus fertile material concentrations of the order of 300-350 g/liter. At these concentrations, solutions of fuels less than ~3% in enrichment ordinarily will be safe because of the low concentrations of fissile material. As enrichment increases above 3%, poison control permits significantly greater throughput for a given plant. Manowitz (38) has estimated that, for a 1-ton/day central processing plant operating on fuel of enrichments greater than 3%, the separation cost with the use of concentration control will be 45-80% greater than for the same plant using predominantly soluble poison control.

8.0 EFFECTS OF A WATER-MODERATED NUCLEAR EXCURSION

IN A SHIELDED RADIOCHEMICAL PLANT

An ORNL study of the types and possible effects of credible accidents in radiochemical facilities has resulted in the establishment of ORNL containment specifications, which are designed to limit the effects of the maximum credible accident to the involved facility. Hazards evaluations (39, 40) have shown that the effects of a water-moderated nuclear excursion, as well as possible chemical explosions, in such a facility would result in minimal personnel exposures and downwind ground contamination.

In view of the tolerable effects of credible nuclear excursions in a "contained" facility, it appears that such accidents should not be avoided at all costs but should be minimized through the use of conditioned control, to a level such that the cost of lost production time,

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Fig. 9. Cost per kilogram of U^{235} of H_3BO_3 , $Cd(NO_3)_2$, Gd_2O_3 , and Sm_2O_3 required to cause $k_{\infty} = 1$ for natural water solutions of poison and U^{235} .

decontamination, and bad publicity resulting from an accident is balanced by the cost savings and other benefits of the use of the conditional controls. Such an evaluation cannot be performed rigorously but must be in the form of an educated judgment. Perhaps it could be decided that the effects of a nuclear accident could be tolerated once per 100-1000 years of plant operation. Such a low probability could be ensured by providing two or three independent controls that are 99 to 99.9% reliable in each of the potentially hazardous operations in a plant.

8.1 Radiochemical Facility Containment Criteria

A schematic diagram of a radiochemical facility which meets the ORNL containment design criteria is shown in Fig. 10. A vessel is in a process cell, which is completely surrounded by a building. The cell, which constitutes primary containment, is capable of withstanding the blast effects of the maximum credible explosion without rupture and permits only a minimum leakage of radioactive material to the secondary containment shell, the building structure. Other criteria for the process vessels, cells, and buildings are:

Filters are to be located such that they will be protected from the maximum credible explosion.

Process vessels are maintained at a vacuum of at least 2 in. w.g. during normal operation by a vessel off-gas system which provides for gas passage through a local scrubber and filter system as well as plant treatment system before being exhausted at a stack.

A cell is maintained at a vacuum of at least 1 in. w.g. during normal operation. The cell ventilation exhaust capacity is at least equivalent to 1/10 of a cell volume per minute. The air intake to the cell is through a roughing filter and check valve. The cell exhaust passes to a cell ventilation manifold, roughing and absolute filters, and from thence to the stack. The cell is sealed such that the leak rate is less than or equal to 1/100 of a cell volume per minute at 2 in. w.g. differential pressure.

The building is maintained at a few hundredths of an inch w.g. vacuum during normal operation. The intake is through duct filters and check valves, and the exhaust is through roughing and absolute filters located at the roof of the building or at the stack. The cell

Fig. 10. Schematic diagram of minimum containment provisions for radiochemical facilities.

ventilation blower must have sufficient capacity to evacuate the building to 0.3 in. w.g. vacuum in 20 seconds by closing the intake. The building is sealed such that a leak rate of no more than 6×10^{-3} building volume per minute will occur at a differential pressure of 0.3 in. w.g. This criterion is included to ensure that the building vacuum will be capable of balancing a vacuum of 0.3 in. w.g., which could be created on the lee side of a building by a 30-mile/hr wind. At ORNL it is pertinent to assume that winds of speed greater than 30 miles/hr are sufficiently rare as to be incredible.

8.2 Magnitude of Credible Nuclear Excursions

The most serious accidents that may credibly occur in large radiochemical facilities are chemical and nuclear explosions that rupture vessels filled with radioactive process solutions. It is currently believed that a radiochemical facility can be designed in such a manner that the maximum credible explosion would generate not more than 100 cu ft of gas and would have blast effects less than that of 3 lb of TNT. Three pounds of TNT liberates approximately 5700 Btu of energy, generates approximately 100 cu ft of hot gases, and creates a shock wave that has a pressure of ~800 lb/sq ft and an energy of approximately 230 ft-lb/sq ft at a distance of 15 ft. Thick concrete cells of the type used in ORNL radiochemical facilities can withstand such explosive effects without rupture.

The magnitude of credible chemical explosions is controlled within the reference TNT detonation by limiting the quantity of reactants in process vessels.

Experience and calculations (41-43) indicate three factors that are pertinent to the maximum nuclear explosion: (1) The most powerful burst will probably be the initial burst. (2) The maximum initial burst will probably be limited by gas generation and thermal expansion to the order of 10^{12} fissions per cubic centimeter (approximately 10^{18} fissions for vessels of approximately 1000 liters capacity, typical of those in moderate-scale plants). (3) It is not incredible that such a burst could rupture a vessel. If the nuclear reaction is not shut down in the initial burst by rupture of the vessel, it is probable that the vessel would not be ruptured at all, and the reaction would recur with

10^{19} - 10^{20} or more fissions until the solution was boiled down or the fissile material forcibly ejected. This type of nuclear accident would have less serious consequences than a burst that ruptures a vessel and scatters a highly radioactive process solution. A nuclear burst of 10^{18} fissions releases approximately 30,000 Btu of energy, 4 cu ft of radiolytic gas, and not more than 100 cu ft of steam. More steam formation would constitute an extremely efficient shutdown mechanism and limit the reaction to less than 10^{18} fissions. Also, since a nuclear burst in solution occurs in millisecond time rather than microsecond time as in TNT detonations, the shock waves will be less energetic than those from the reference TNT detonation.

It should be noted that the number of fissions in the usual criticality excursion that does not rupture a vessel but tends to recur over a relatively long period of time may be decreased by the use of a built-in shutdown mechanism. Pressurized tanks, filled with concentrated cadmium solutions, which would automatically dump their contents into preselected vessels in the event of an abnormal neutron background, have been suggested for this use in shielded cells at ORNL.

8.3 Effects of Credible Nuclear Excursions

The maximum chemical explosion would cause the formation of an aerosol of the radioactive material in the cell air, and a small fraction of the aerosol would reach the environment through the vessel and cell off-gas systems and through successive leaks from the cell and the building. The maximum nuclear burst would disperse new gaseous fission products in addition to the aerosol, and operating personnel would receive direct prompt gamma and neutron radiation through the shield.

The prompt gamma and neutron dose that would be received through a cell wall from a nuclear burst of 10^{18} fissions is shown in Table 4. From these data one may conclude that the direct radiation constitutes only a few millirem and is not a significant hazard in a typical radiochemical facility in which the concrete walls are 5 to 6 ft thick. It would not even be significant in a recurring nuclear reaction, since personnel could be expected to evacuate the area of the shield before more than the order of 10^{18} fissions occur. Lethal doses of gamma and neutron radiation could be received when there is little or no

Table 4. Prompt Neutron and Gamma Dose at Exterior of a Normal Concrete Shield from a Nuclear Reaction of 10^{18} Fissions^a

Concrete Shield Thickness, ft	Dose at Outer Side of Shield, rem	
	From Metal Nuclear Reaction	From Nuclear Reaction in Aqueous Solution
1	88,000	5,200
3	317	23
4	17.0	1.9
5	0.960	0.14
6	0.059	0.012

^aThe dose rate may be calculated for any other number of fissions by a direct proportion.

Table 5. Effects^a of a Nuclear Excursion of 10^{18} Fissions that Ruptures a Storage Tank Containing 60 kg of Pu Solution at 100 g/liter in a Bldg. 3019 Process Cell

	Dose, rem	
	From F.P.'s	From Aerosol of Pu and Nonvolatile F.P.'s
Maximum downwind dose from vessel off-gas system release	0.22	0.0043
Maximum downwind dose from cell ventilation system release	0.15	0.0035
Maximum downwind dose from successive leaks through the cell and building walls	$< 10^{-6}$	$< 10^{-6}$
Average 2-minute dose to personnel in secondary containment zone	0.18	0.031

^aGround is not contaminated to hazardous level downwind from building or stack.

concrete shielding; this suggests that the use of thick concrete cells should be considered even for operations with fissile materials that are not inherently gamma radioactive.

A calculational model for evaluation of the downwind radiation dose and ground contamination resulting from radioactive gaseous fission product and aerosol release in nuclear and chemical accidents has been developed. The model allows consideration of the properties of containment systems, properties and behavior patterns of radioactive gases and aerosols, efficiency of air cleaning devices, and meteorological dispersion effects. The model and the results of its application to credible accidents in typical radiochemical facilities are described in ORNL reports (39,40).

The results of the radioactive release calculations may best be illustrated by an example. The effects of the maximum credible nuclear accident in Bldg. 3019 are shown in Table 5. The maximum credible nuclear accident in this facility was considered to be an excursion that ruptures a plutonium storage tank containing 60 kilograms of plutonium solution at a concentration of 100 g/liter. The combination ingestion-external radiation doses are controlled by the gaseous fission products, which are not appreciably removed in the ventilation system, rather than by the solution aerosol of plutonium and nonvolatile fission products. In the case of the radionuclides which are predominantly internal radiation hazards, the bulk of the dose does not occur during the exposure period but is accumulated over a lifetime because of the presence of the radionuclide in the body. The maximum downwind dose because of successive leaks through the cell and building wall is insignificant compared to the approximately 200-mrem dose from activity released through the filtered vessel and cell ventilation systems. The average dose to operating personnel due to activity forced from the cell into the secondary containment shell while the cell is pressurized is not expected to exceed 200 mrem. The ground downwind from the building and stack would not be contaminated within 10% of the permissible level.

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