APAE No. 43
Volume I

APPR-1
RESEARCH AND DEVELOPMENT PROGRAM
DECONTAMINATION PROGRAM
TASK II

VOLUME I
CONTAMINATION AND DECONTAMINATION IN NUCLEAR POWER REACTORS

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APAE NO. 43
Vol. I

DECONTAMINATION PROGRAM

TASK II

VOLUME I

CONTAMINATION AND DECONTAMINATION IN NUCLEAR POWER REACTORS

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ABSTRACT

A survey of the problem of reactor system contamination by radioactive material and methods that have been employed to remove the material was carried out. Following this survey, an investigation of chemical solutions was undertaken to find one which might be successfully employed in the decontamination of a stainless steel steam generator. From a preliminary screening, the most promising chemical method from the view point of minimum corrosion and maximum decontamination is a caustic permanganate treatment followed by an acid rinse.

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1.0 INTRODUCTION

The removal of radioactive deposits from work surfaces and equipment is a major problem wherever radioactive materials are encountered. The technical literature on the subject, however, deals primarily with methods and procedures for laboratory or manufacturing facilities. (1, 2) However, the problem is encountered in the building and operation of nuclear reactors and nuclear chemical processing plants, and recently a considerable effort has been devoted to understanding radioactive contamination, its buildup, and subsequent removal. The cost of nuclear power plants as well as their ease and simplicity of maintenance depends upon whether effective means can be found to control radioactivity buildup or to cheaply and simply decontaminate the plant.

This report is the first in a series on the Decontamination Program carried out by Alco Products, Inc., in conjunction with the operating contract for the Army Package Power Reactor (APPR-1), a pressurized water reactor located at Fort Belvoir, Virginia. Subsequent reports will include the results of extensive loop testing of chemical decontamination solutions and the recommended procedure for APPR-1 decontamination.

2.0 OBJECTIVE

The results of a survey of the more important aspects of nuclear reactor contamination and decontamination are presented in this report, as well as the results of screening of chemical decontamination solutions investigated under the APPR-1 Decontamination Program.

3.0 SUMMARY AND CONCLUSIONS

The buildup of activated corrosion products on the primary system surfaces of nuclear reactors may present a serious restriction to the normal operation of the installation. Continued buildup of this activity can seriously restrict accessibility and prevent necessary maintenance. As a consequence, rapid and efficient decontamination is required to reduce the plant or system to safe working levels.

The corrosion product deposits are composed chiefly of magnetite (Fe_3O_4) with small amounts of nickel and chromium oxides. The radiochemical composition depends upon the materials of construction; Fe^{59}, $Co^{60, 58}$, Mn^{54} and Cr^{51} have been identified in APPR-1 corrosion product scale.

The extent of contamination by activated corrosion products is influenced by many factors including water chemistry, time, temperature and/or pressure, precorrosion treatment, and the type of metal involved.

Ordinarily a tenacious radioactive film is formed which is difficult to remove even by the action of strong chemical agents.

Considerable effort has been devoted to the removal of corrosion product and fission product activity as well as uranium oxide from metal surfaces. The techniques that have been employed include chemical removal, ultrasonics, and eletrolytic descaling. However, most of the work concerned with the decontamination of large facilities or inaccessible components has involved chemical methods. A reducing complexing solution ("RDS"), developed by the Westinghouse Atomic Power Division, has been employed to remove activated corrosion product scale from a number of facilities including the naval reactor prototype (S1W). The Turco "4501" Process has been used to decontaminate the KAPL 120 Loop at Hanford Atomic Power Operations on two separate occasions. A chromous sulfate - sulfuric acid has been used to decontaminate the homogeneous reactor experiment. Generally, chemical decontamination of nuclear facilities has achieved only partial success. At this time, however, chemical decontamination seems to represent the most practical way of decontaminating large facilities.

Preliminary screening results obtained in the APPR-1 Decontamination Program indicate that a caustic permanganate-rinse treatment may be an effective chemical method for removing activated corrosion product scale. Decontamination factors of 20 or more have been obtained. The available corrosion information indicated no evidence of intergranular corrosion. However, an extensive evaluation of this particular chemical treatment is necessary before application to a large reactor facility can be considered.

4.0 DISCUSSION OF RESULTS

4.1 The Problem

4.1.1 Activity Buildup in Pressurized Water Reactors

4.1.1.1 Naval Reactor, PWR Experience

The need for periodic decontamination of nuclear facilities is the result of the radioactive deposit or film that tends to build up on system components. Prolonged buildup can seriously hinder necessary maintenance and thereby restrict operation of the facility. Considerable information is available in the unclassified literature concerning the characteristics, the magnitude, and the attempts that have been made to predict theoretically the radioactivity buildup in pressurized water reactors (3). More recently Culver (4) and Meem (5) have used system parameters and early results from the Army Package Power Reactor (APPR-1) to obtain specific information regarding activity buildup in the Nuclear Merchant Ship Reactor.

4.1.1.2 Army Package Power Reactor (APPR-1) Experience

Radiochemical analysis of circulating water and particulate matter in the APPR-1 has indicated the presence of the long-lived species $Co^{60, 58}$, Fe^{59}, Mn^{54}, and Cr^{51} (6-8). Practically all of this activity except manganese is associated with the particulate matter. A slow but steady rise in activity has been observed in the APPR-1. The greatest contribution to the activity is from Co^{60} and Co^{58}. The former arises from an n-γ reaction on the natural cobalt present as an impurity in stainless steel. The Co^{58} arises from an n-p reaction on the Ni^{58} which is present in the stainless steel throughout the system. Since the reactor went critical in April 1957, the steam generator head has been lowered on two separate occasions to permit an examination of the interior and to collect dose rate information. The first occasion was in July 1957, immediately following a 700 hour full power test; the second occasion was in November 1958. The dose rates in the steam generator on these two occasions are listed on Page 6.

Dose rates at the end of core life (November 1959) are estimated to be 25-30% higher than November 1958 values. No appreciable contribution due to fission product activity is anticipated, although some fission products are present. (9)

TABLE 1 APPR-1 Steam Generator Gamma Dose Rates

Location	July 1957 700 E.F.P.H.*	November 1958 7700 E.F.P.H.*	Instrument Used
A. 12" below tube sheet	280 nr/hr.	-	"Cutie Pie"
B. Tube Sheet, Inlet		6.3 R/hr.	Jordan AG-50 BP Meter (unshielded)
Tube Sheet, Outlet (next to vertical baffle plate)		4.3 R/hr.	
Tube Sheet, Outlet -side		2.9 R/hr.	

* equivalent full power hours

4.2 Decontamination Philosophy

The primary function of decontamination is to facilitate maintenance and emergency repair of the plant facility. The plant may be a nuclear power plant or a nuclear fuel reprocessing plant. Ease of maintenance is of particular importance in the small power plants where staffs are small and excessive dose rates could seriously restrict operation.

The Westinghouse Atomic Power Division and the Oak Ridge National Laboratory have devoted considerable time and effort to the decontamination of nuclear facilities. While different in the type and extent of the radioactivity involved, the goal at both installations has been the same, i.e. achieving high decontamination factors with low corrosion rates, short exposure times, and low cost.

The naval reactor program has been concerned with the activity buildup in naval reactors. In December 1957, the naval prototype (S1W) at Idaho Falls, Idaho, was decontaminated using a concentrated reducing solution ("RDS") in a feed and bleed type of application. The results of this operation are discussed in a subsequent section of this report. While the type of activity encountered in S1W is different from that found in the APPR-1 (due to differences in cladding material), the general behavior and mechanisms involved are probably the same.

The ORNL program has been considerably more varied than the naval reactor program because of the numerous facilities requiring decontamination. Components containing homogeneous reactor fuels together with reprocessing equipment have been decontaminated.

It is generally felt that the frequency of decontamination should be dictated by the economics of maintenance. It is essential to determine the dose rate which will permit maintenance to be performed with the smallest staff and in the shortest possible time. This necessitates the continuous checking of dose rates at numerous points on the reactor facility (S1W has more than 150 such points)(10). This information must be subject to constant review in order to determine the location of "crud traps" and to anticipate future activity levels.

In summary then, a successful decontamination program must fulfil the following requirements:

1. <u>Knowledge of the type of radioactive contamination involved together with its rate of buildup.</u>

   This would include chemical and radiochemical compositions, relative adherence to the material surface, and a probable mechanism for its buildup.

2. <u>The decontamination treatment must be capable of achieving maximum removal of activity.</u>

   The chemical decontaminating solution must not result in excessive corrosion nor deleterious metallurgical effects. The subsequent corrosion rates and buildup of activity must not be increased as a result of the decontamination treatment. The solution must be stable at high temperatures, pressures and radiation levels. Excessive decomposition could give rise to gas formation and result in pump cavitation.

3. <u>The decontamination treatment must be applied in a manner that is simple, brief, and economical.</u>

4. <u>Adequate provisions for waste disposal.</u>

   There must be suitable equipment to process large volumes of highly contaminated liquids.

5. Recommendations for future reactor designs.

Future reactors must be designed with the problems of decontamination in mind and in the light of recommendations resulting from decontamination studies. This would include any provisions for isolation of contaminated components, removal of stagnant areas where deposits may collect ("crud traps"), and the design and location of decontamination and waste disposal equipment.

4.3 Nature of the Activated Corrosion Product Scale

4.3.1 Comparison of Chemical and Spectrographic Compositions.

The scale encountered in pressurized water reactors is composed chiefly of magnetite (Fe_3O_4) with smaller amounts of nickel and chromium oxides. Table 2 lists the corrosion product scale ("crud") composition as found in naval reactors employing Zircaloy clad fuel elements.

TABLE 2 Chemical Composition of Activated Corrosion Products in Reactors Employing Zircaloy Clad Fuel Elements (11)

Element	System Degassed Flow Rate 6 Ft/Sec	System Hydrogenated 6 Ft/Sec	System Hydrogenated 27 Ft/Sec
Iron	86%	60%	75%
Nickel	6.4%	6.7%	6.3%
Chromium	1.8%	3.1%	13%

These compositions are in the same range as found in the Army Package Power Reactor (Table 3). This reactor, employing stainless steel clad fuel elements (Type 304), is essentially a hydrogenated system, with a flow rate of 10 Ft/Sec.

TABLE 3 Chemical Composition of APPR-1 Activated Corrosion Products (9)

Element	Average Percent	Range Percent
Iron	52	30-67
Nickel	5.0	0.7-7.9
Chromium	<0.32	<0.002-0.9
Manganese	<0.25	<0.006-0.6
Cobalt	<0.17	----

Spectrographic analysis has revealed, in addition to the elements listed in Table 3, the presence of small amounts of silicon, aluminum, molybdenum, silver, and magnesium(9).

4.3.2 The Effect of Temperature on APPR-1 Corrosion Scale

The activated corrosion product scale that is found on the primary system surfaces of pressurized water reactors can be extremely tenacious. The tenacity depends upon both the operating conditions and the length of exposure. This is shown by the data of Table 4 where contaminated Type 304 stainless steel metal coupons were continuously electrolytically descaled for periods totaling 60 minutes. (Fig. 1). The samples were identical in size (3-1/2 x 1/2 x 1/16-inch), heat treatment, and surface finish and have been exposed up and downstream of the APPR-1 primary blowdown cooler for approximately 4-6 months. As a result, some samples were exposed to APPR-1 primary system conditions while others were exposed to the same coolant at reduced pressure and temperature. The samples to be descaled were made cathodic to a lead anode in a solution of 5% sulfuric acid and 1 gram/liter of thiourea. A current density of 1.3 amps/in^2 was used. At specified times, the samples were removed from the descaling solution, rinsed with distilled water, and the gamma activity determined by a Geiger Muller tube. The results indicate that the samples exposed to the APPR-1 system conditions (430°F at 1200 psi) possessed a much more tenacious scale than samples exposed to 100°F at 45 psi. Approximately 7-8% of the gamma activity remained after 60 minutes on the metal specimen exposed to the higher temperature and pressure. This activity may have been very tightly adsorbed on the metal surface or even diffused into the base metal. Ultrasonic cleaning of Type 304 stainless steel test coupons has also indicated the same temperature and/or pressure effect (9).

TABLE 4 Results of Continuous Electrolytic Descaling Type 304 Stainless Steel 3-1/2 x 1/2 x 1/16-inch Coupons

Blowdown Conditions	No. Samples	Percent Gamma Activity Removed at Indicated Times (Minutes)				
		5	10	20	30	60
Upstream of Cooler 430°F at 1200 psi	4	70	80	84	87	92
Downstream of Cooler 100°F at 45 psi	2	99	100	100	100	100

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Fig. 1 Laboratory Bench Facilities For Dynamic Autoclave and Electrolytic Descaling Testing

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Figure 2 is a photo of a section of the APPR-1 stream generator flow separator. This section has been exposed in the APPR-1 primary system for approximately 11,500 hours. Attempts to remove the black, lustrous scale by electrolytic descaling were unsuccessful. It appears, therefore, that temperature and/or pressure, together with exposure time, tend to fix the deposited activity more tightly on a metal surface.

4.3.3 Metal Differences of APPR-1 Corrosion Scale.

The accumulation of activity on the metal test coupons is also dependent upon the material involved. Brown et al (8) have shown that, for equal exposure times to APPR-1 reactor coolant, Babcock and Wilcox Croloy 16-1 stainless steel accumulated the most activity followed by carbon steel and Type 304 stainless steel. It was found that the Croloy 16-1 accumulated 3-6 times as much activity as the Type 304 stainless steel. Furthermore, the Croloy 16-1 exhibited the most adherent film while carbon steel exhibited the least adherence. On the basis of this data, it might be expected that Croloy 16-1 would be the most difficult to decontaminate. However, later studies in connection with APPR-1, indicated this is not necessarily true.

4.3.4 Probable Mechanisms for Buildup of Activity

4.3.4.1 Deposition and Atom Exchange

A number of mechanisms have been cited to account for the accumulation of activity on reactor surfaces. A discussion of these mechanisms is given in reference (3). Deposition and atom exchange are covered in detail together with the evidence supporting each theory. An electrochemical-exchange type of mechanism has also been proposed to account for some of the more recent observations in the APPR-1 (7). If atom exchange or diffusion of activity were the basic mechanism involved in activity buildup, it would be extremely difficult to remove the radioactive contamination. Under these circumstances the radioactivity would no longer be merely on the surface, but would actually have become part of the base metal.

4.3.4.2 Adsorption Mechanism for Activity Buildup

There is some evidence to indicate that the accumulation of activity on metal surfaces may involve physical or chemical adsorption. Here the force of attraction between the metal surface and the contaminant is supplied by the unsaturated molecular forces at the metal surface. These forces may be small, i.e. 10 kcal (physical or Van der Waal adsorption) or large, i.e. 50-100 kcal (chemical or activated adsorption). (12) WAPD has estimated the force of attraction between certain fission product contaminants and metal surfaces to be approximately 30 kcal. (13).

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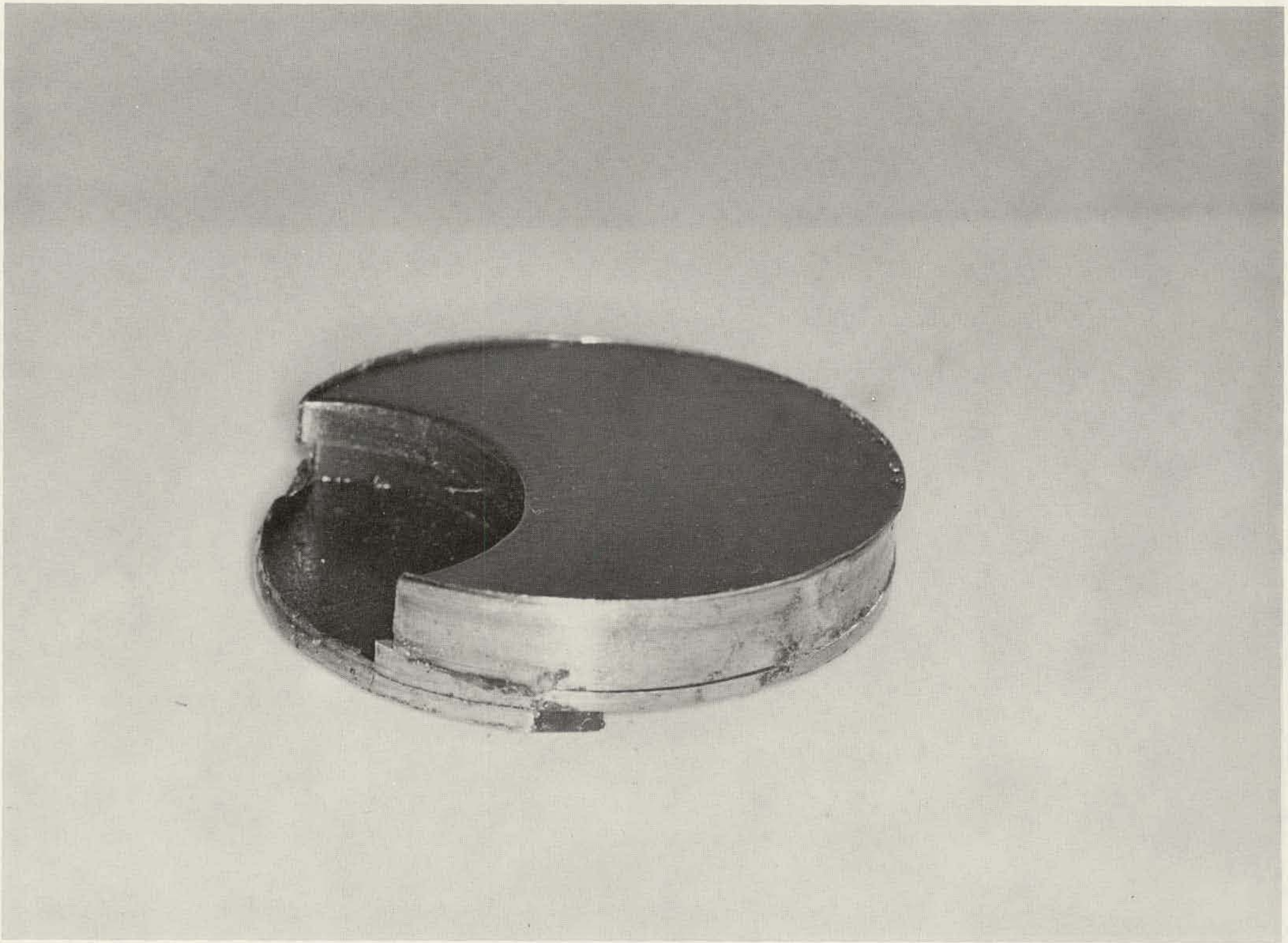

Fig. 2 Section of APPR-1 Steam Generator Inlet and Outlet Dividing Plate After 11,500 Hours Exposure

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Campbell (14) has shown that the contamination of Type 347 stainless steel surfaces by "Purex" process waste solutions follows the Freundlich adsorption equation. The relationship is given by

$$Y = k\,C\,1/n$$

where Y = activity per unit area

C = equilibrium concentration (activity per unit volume)

n, k = empirical constants

It was surprising that the data did not fit the Langmuir adsorption equation as the two relationships can be used almost interchangeably when measuring the adsorption of gases on solids.

The contamination of Type 304 stainless steel surfaces by both APPR-1 activated corrosion products and primary coolant was investigated. The corrosion products were dissolved with a mixture of nitric, perchloric, and hydrofluoric acids and brought to a pH of 2 with a mild caustic solution. Successive dilutions were made of this solution and 200 cc aliquots of each were used in the contamination experiments. The metal coupons (1-3/4 x 1/2 x 1/16-inch) were in the annealed condition and all possessed the same surface finish (30-35 RMS). These were suspended in beakers containing the active solutions for periods up to 188 hours. At intervals during the exposure the samples were removed, rinsed with distilled water, and then counted with a thin mica end window Geiger Muller tube. The samples were then returned to the solutions for additional exposure. The variation of activity in counts per min per coupon as a function of exposure hours is given in Table 5.

TABLE 5 Contamination of Type 304 Stainless Steel at 25°C APPR-1 Dissolved Corrosion Products (pH=2)

Solution No.	Solution Activity Counts/cc	Average Counts/Minute/Coupon After Indicated Exposure Hours									No. Samples
		1	2	3	4	18	25	43	88	188	
1	2,714	205	-	-	333	-	436	-	515	-	4
2	6,411	343	-	420	-	-	522	-	595	-	4
3	14,315	749	-	-	-	924	-	994	-	1012	2
4	20,042	744	891	989	-	1082	-	1140	-	1155	2

The curve in Fig. 3 indicates that the equilibrium activity was adsorbed in the metal coupons within 88 hours. More than 85% of this activity was adsorbed within the first 24 hours. A plot of the equilibrium activity after exposure times of 24 and 88 hours versus solution activity indicates agreement with the Freundlich adsorption equation (Figure 4).

A similar experiment was performed on Type 304 stainless steel (as annealed and as sensitized*) using APPR-1 primary coolant water which had been concentrated from 10 liters to approximately 500 cc. Practically all the activity in the coolant is associated with suspended insoluble corrosion products ("crud"). It is interesting to note that the relationship between dissolved activity and occluded activity also follows the Freundlich adsorption equation (Figure 5). The sensitized Type 304 stainless steel accumulated about 1.4 times as much activity as the coupons in the annealed condition. Unpublished data obtained as a result of exposing Type 304 stainless steel to the APPR-1 primary system indicated that the sensitized specimens accumulated about twice the activity as the annealed specimens. Corrosion rates were also in the same ratio.

There appears to be strong evidence that the contamination of metals by dissolved or suspended activated corrosion products involves a physical or chemical adsorption of the activity on the metal surface. However, the effects of temperature, pressure, corrosion rate, water chemistry, and surface condition remain to be investigated. Assuming a Langmuir or Freundlich type of adsorption, increasing the temperature would tend to reduce adsorption, while increasing pressure or surface area would increase it. Waldman (15) has shown that surface finish is not as important a variable as position of the metal surface with respect to the reactor coolant. It was pointed out that there was a greater difference in activity uptake between samples having different precorrosion times than between surface finishes (Table 6).

TABLE 6 Activities of Precorroded Autoclave Coupons

	LiOH Solution							
	Without H_2				With H_2			
Precorrosion Time - Hrs.	100		200		100		200	
Surface Finish (avg. RMS)	30	125	30	125	30	125	30	125
Sample Activity(avg.(cpm)	489	641	1449	1446	1368	1694	1860	2532
Max.	594	764	2004	1991	1684	2064	2742	3027
Min.	411	482	1252	1198	988	1300	1272	2136

* Sensitized at 1200°F for 10 minutes; air cooled.

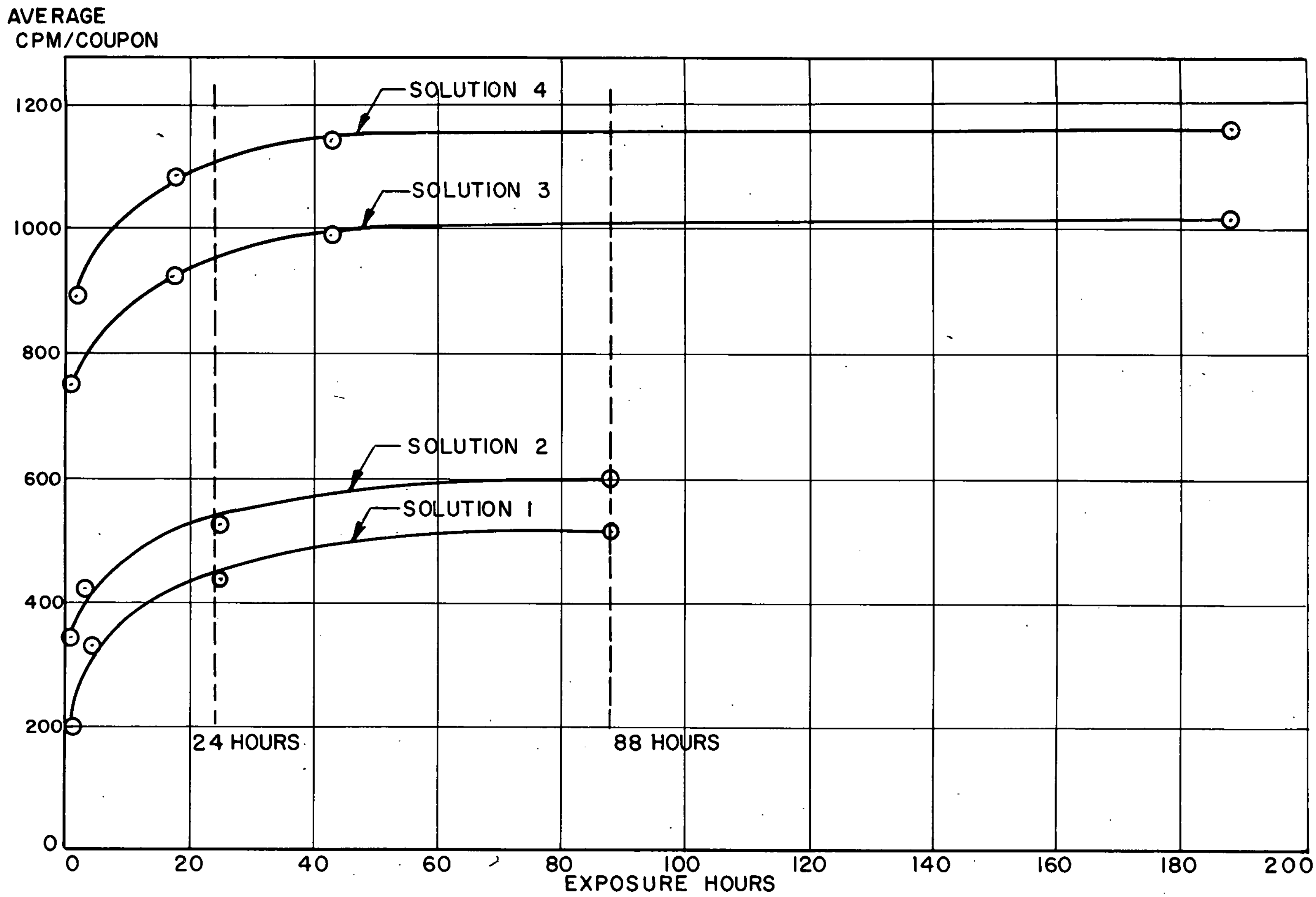

Fig. 3 Contamination of Type 304 Stainless Steel at 25° C
By APPR-1 Dissolved Corrosion Products (pH = 2)
Effect of Exposure Time

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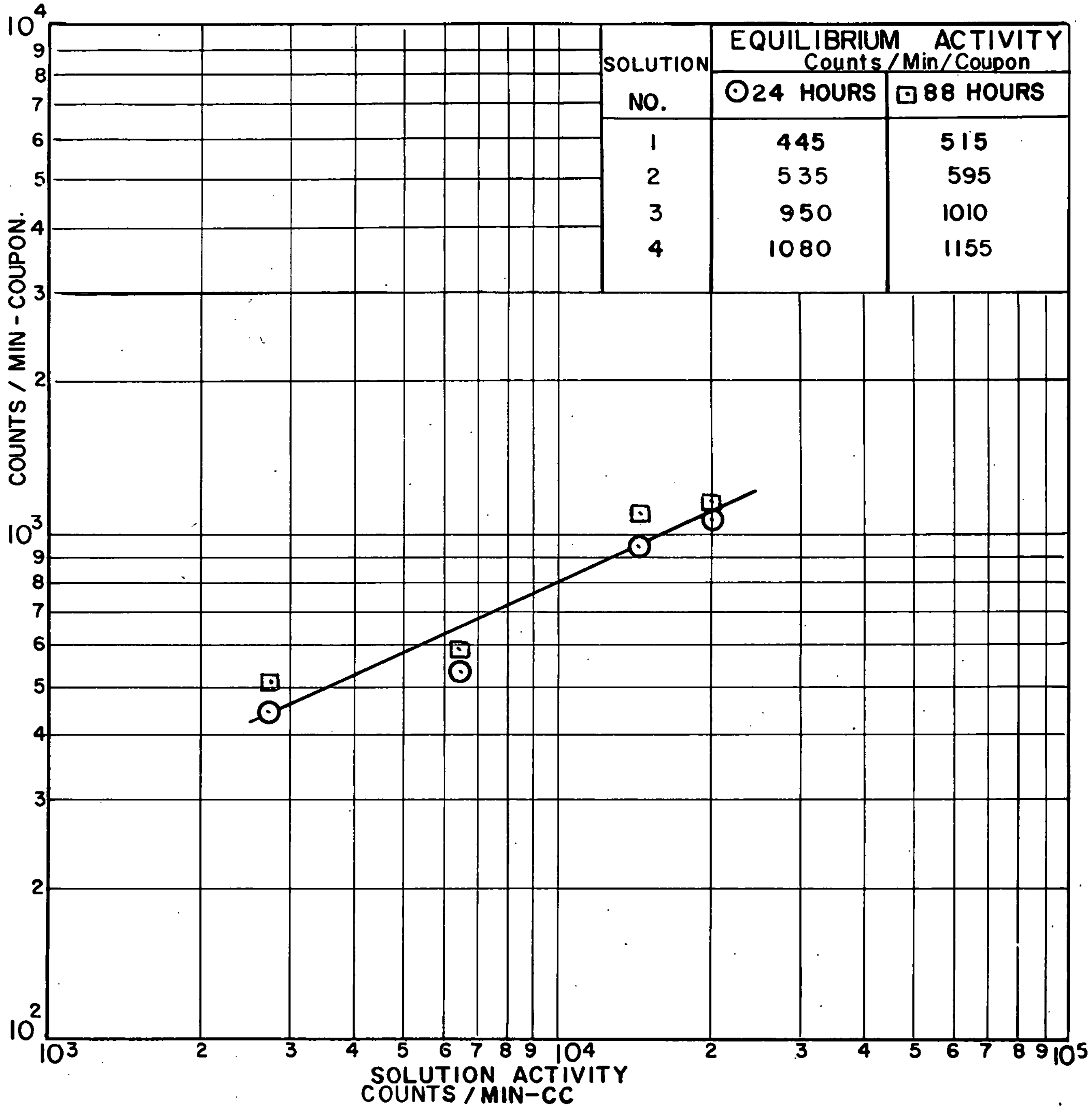

SOLUTION NO.	EQUILIBRIUM ACTIVITY Counts / Min / Coupon	
	⊙ 24 HOURS	⊡ 88 HOURS
1	445	515
2	535	595
3	950	1010
4	1080	1155

Fig. 4 Contamination of Type 304 Stainless Steel at 25° C By APPR-1 Dissolved Corrosion Products (pH = 2) Test of Freundlich Adsorption Equation

$$\text{Log } Y = \log k + 1/n \log C$$

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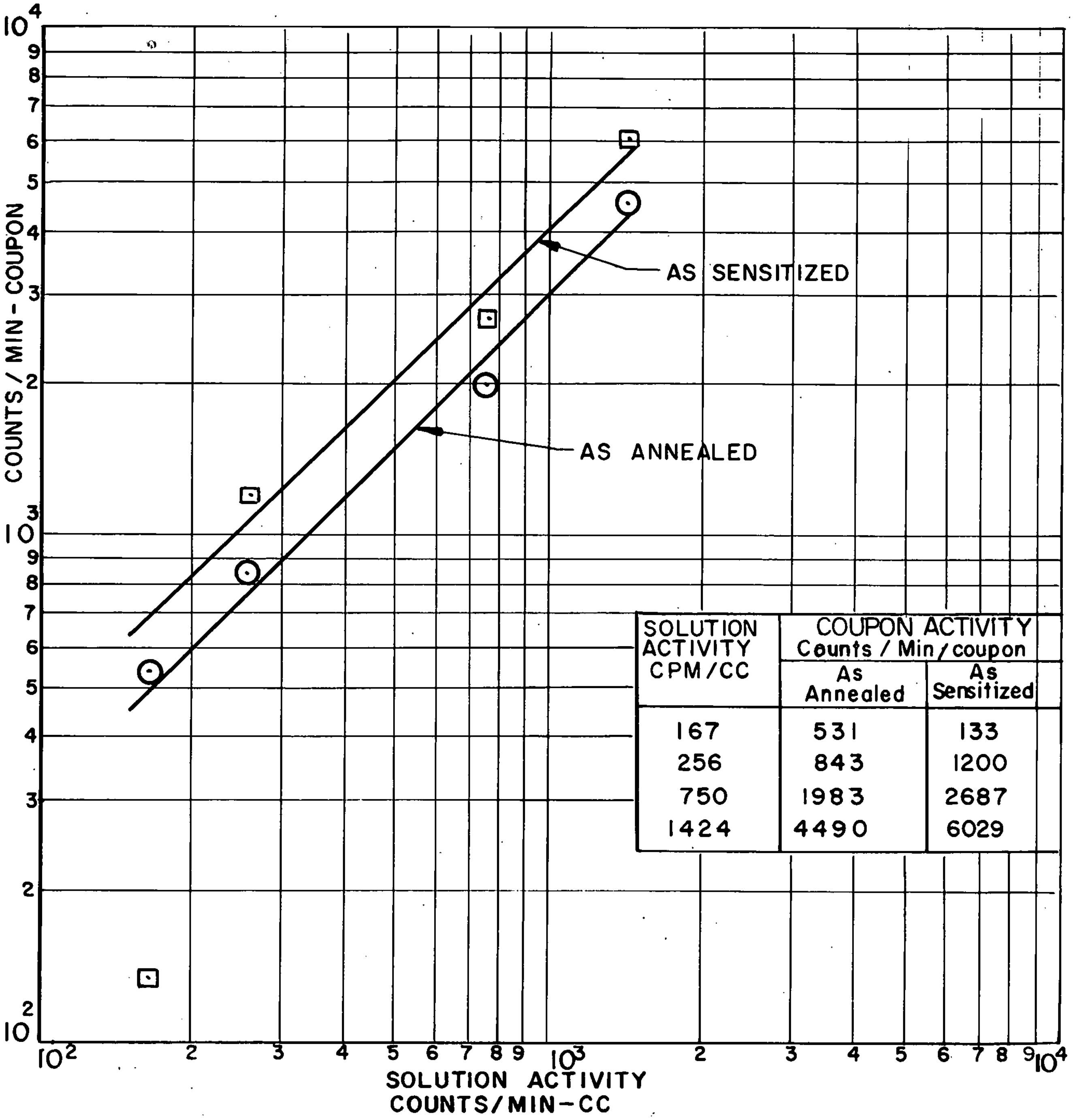

SOLUTION ACTIVITY CPM/CC	COUPON ACTIVITY Counts / Min / coupon	
	As Annealed	As Sensitized
167	531	133
256	843	1200
750	1983	2687
1424	4490	6029

Fig. 5 Contamination of Type 304 Stainless Steel at 25° C By APPR-1 Primary Water (pH = 7) For 24 Hours Test of Freundlich Adsorption Equation

$$\text{Log } Y = \log k + 1/n \ \log C$$

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4.4 Methods for Decontamination

4.4.1 Chemical Methods

At the present technological stage of reactor decontamination, the most feasible method appears to involve the use of chemical agents. These chemical agents are designed to remove the corrosion product activity, fission product activity and any fissile fuel that may be present in the reactor system as a result of a defective fuel element.

4.4.1.1 Methods for Application of Chemical Solutions (Feed-Bleed vs. Fill-Flush)

There are two general methods for accomplishing chemical decontamination. These are the feed-bleed and the fill-flush methods. Each method has its basic advantages and disadvantages and consequently the choice of a method depends upon the system under consideration(16). A schematic for a decontamination system using either method is shown in Fig. 6.

With a feed and bleed type operation, a constant supply of fresh decontamination solution is introduced into the system while a part of the exhausted solution is bled off. The amount of solution fed is fixed at an amount necessary to maintain the following material balance:

Total Feed = amount needed for reaction + amount decomposed by thermal effects + amount decomposed by radiation effects.

Once this concentration is established, it can be maintained very simply by adjusting the bleed rate.

With a fill-flush operation, the system to be decontaminated is filled with the solution which is circulated through the system with no additional feed during any one cycle. With this type of operation, the initial fill must be of such a concentration as to allow for depletion of the reagents during the cycle.

With a feed-bleed operation, a close control of the concentration can be maintained because of the constant bleed of a portion of the total system volume. This permits a continuous check of the concentration and the radioactivity in the system. A change in the concentration can be accomplished by simply changing the bleed rate which correspondingly changes the feed rate. This type of direct control is not possible with a fill-flush system because (a) the concentration of the loop solution is not constant, but changes as the solution is exhausted, (b) if a sample

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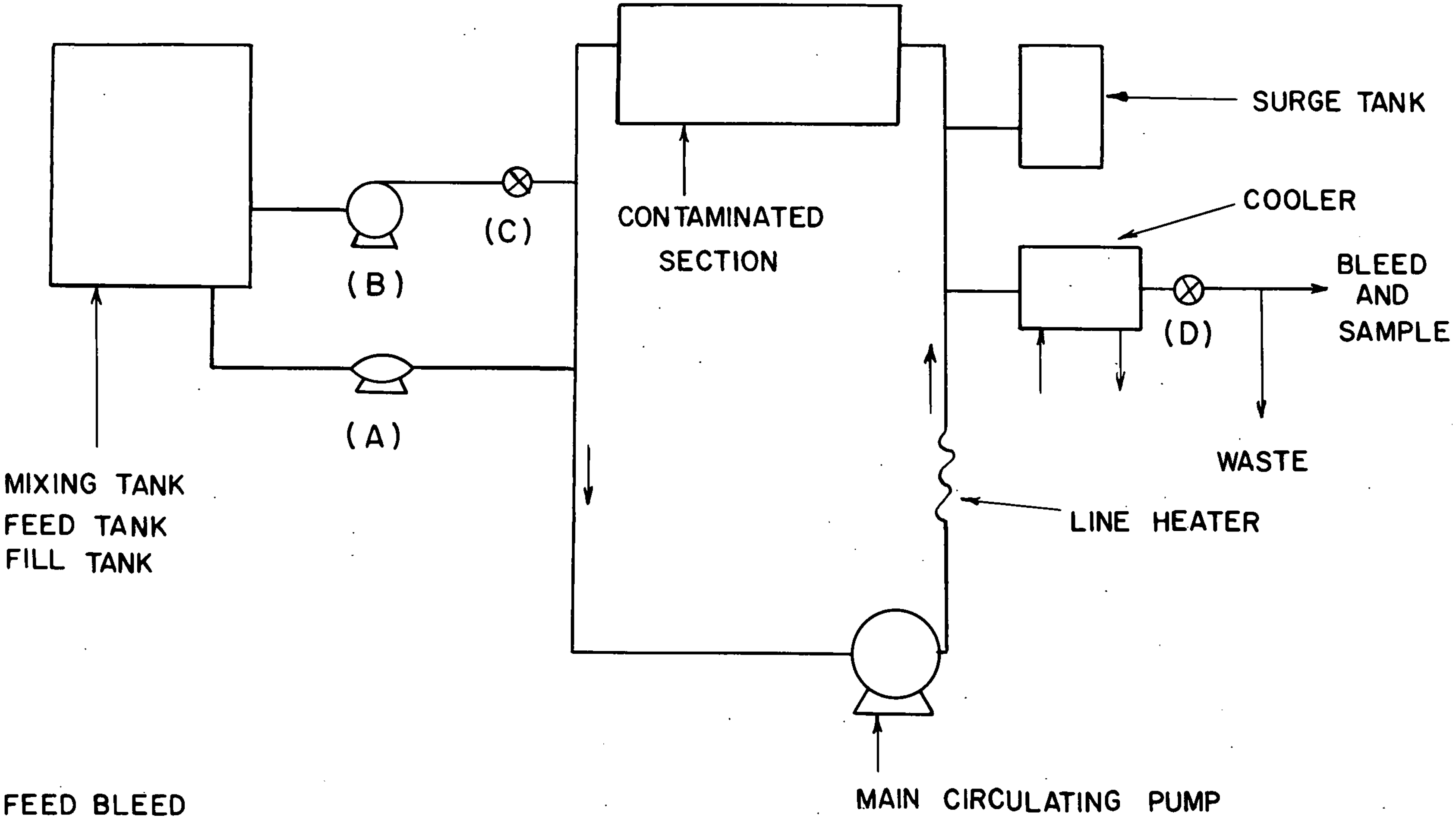

FEED BLEED

USE POSITIVE DISPLACEMENT TYPE
PUMP (A) FOR OPERATION

FILL FLUSH

USE CENTRIFUCAL TYPE PUMP (B) FOR FILLING
CLOSE VALVES (C) & (D) FOR OPERATION

Fig. 6 Equipment Schematic For Feed-Bleed or Fill-Flush Decontamination Procedure

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is withdrawn from the system, fresh solution must be introduced and this may change the system chemical balance. Decontamination by a fill-flush operation requires a higher initial concentration which could result in a higher corrosion rate than in feed-bleed operation. In most systems, there will probably be so-called "dead legs" where some of the decontamination solution remains after the loop is drained. It is imperative that this solution be removed to permit further reactor operation and to prevent excessive corrosion. This can be accomplished by flushing the system until the drain water is at the correct purity. With a fill-flush operation, the higher concentration of solution in these "dead legs" or "pockets" would require more flushing cycles than a feed-bleed operation.

The fill-flush method of reactor decontamination may be advantageous when corrosion rates are not excessive and costs must be kept to a minimum. If the system pump is used for circulation, the only additional equipment required is a pump for introducing the chemical solution into the reactor system together with draining and waste storage facilities. It may be possible to adapt the drain facilities of the reactor for decontamination purposes.

An evaluation of all the factors, i.e. corrosion rates, solution characteristics, radiation levels, and required engineering modifications, will determine the most suitable method for a particular system. The application of these methods in specific cases is discussed later in this report.

4.4.1.2 Parameters for Decontamination

The correct temperature for a chemical decontamination depends upon many factors. There is a required minimum temperature to maintain the solubility of the chemical agents being used. Higher temperatures tend to increase both corrosion rates and the rate of scale removal. Some solutions tend to decompose at higher temperatures (17). A particular solution may also require a minimum temperature for the chemical reaction to take place (18). A balance of all these factors is necessary to determine the optimum temperature of a chemical solution. WAPD has fixed the temperature of a reducing complexing solution at 250°F ± 10°F as a compromise between the effective scale removal temperature and low corrosion rate (19, 20).

The optimum pressure for a decontamination operation should be high enough to prevent steam bubble formation and keep any decomposition or corrosion gases in solution. Often, however, it is dictated by the pressure necessary for operating the circulating pumps. The nominal pressure for S1W decontamination was fixed at 300 psi. (19)

This pressure was sufficiently high to keep decomposition and corrosion gases in solution and prevent boiling. It was also convenient for operating the main coolant pumps without danger of cavitation.

In general, the flow rate used in reactor decontamination should be the highest practical velocity. In addition to ensuring proper mixing and heat dispersal, high velocities create a turbulent effect which may enhance the removal of activity from the system surfaces (18).

4.4.1.3 Waste Disposal

The problem of waste disposal is also associated with the decontamination of nuclear facilities. An extensive analysis and discussion of the general problem can be found in Reference 21. Specific problems may be encountered with regard to the chemical decontaminating solutions. These solutions are often-times highly corrosive; consequently, long-term storage may not be feasible. The presence of complexing chemicals such as EDTA (ethylenediaminetetraacetic acid) and citrate may prevent or limit attempts to concentrate and purify the solution by precipitation or ion exchange techniques. The volume of contaminated liquid may be quite large due to the large volume of rinse water that is necessary to bring the system water back to operational purity. Concentrating the waste solution by evaporation might therefore be considered a prelude to any subsequent processing by either precipitation or ion exchange methods. PWR employs a two-stage evaporation process to concentrate waste solutions followed by underground storage of the high activity evaporator bottoms at the site (22). A similar disposal system will be used in the Dresden Nuclear Power Station (23).

4.4.1.4 Full Scale Chemical Decontamination Results

4.4.1.4.1 WAPD 29 Loop (24)

A feed-bleed decontamination operation was performed on the WAPD 29 loop at the Materials Testing Reactor (MTR) in July 1957. This loop, which is fabricated of Type 304 stainless steel, has an out-of-pile surface area of 53 ft.^{2} and a total volume of 15 gallons (10 gallons circulating and 5 gallons in the surge tank). The contamination of the loop was due to: (a) fission product activity, (b) release of UO_2 from the rupture of two fuel wafers, and (c) the buildup of deposited corrosion product scale over "years" of operation. To accomplish the decontamination, two solution treatments were used: (a) an oxidizing - complexing solution to remove UO_2 and fission products (Table 7), and (b) a reducing-complexing solution to loosen and dissolve the corrosion product scale (Table 8).

TABLE 7 WAPD Oxidizing - Complexing Solution

Chemical	Concentration (gms/liter)
Acetanilide	0.5
Ethylenediaminetetraacetic Acid	0.75
Sulfamic Acid	2.5
Hexamethylenetetramine	0.5
Hydrogen Peroxide	1.5

TABLE 8 WAPD Reducing - Complexing Solution ("RDS")

Chemical	Concentration (gms/liter)
Hydrazine	5.0
Ethylenediaminetetraacetic Acid	3.5
Sulfamic Acid	25.0
Hexamethylenetetramine	1.0

The inability to isolate the in-pile section of the loop for the initial oxidizing- reducing solution treatment presented a problem. It was feared that a high residual gamma radiation field in the in-pile section would cause excessive decomposition of the solution. As a consequence, the solution was introduced at the exit of the in-pile section to permit complete circulation of the solution prior to entering the high radiation area. This was coupled with a high bleed rate to replenish the system with fresh solution. The conditions used were:

Pressure	500 psig
Circulation Flow Rate	7.5 gpm
Feed-Bleed Rate	0.67 gpm
Temperature	220°F
Residence Time at Temperature	9 hours, 40 minutes

The second phase of the decontamination using the reducing-complexing solution was accomplished with the in-pile section isolated. Therefore, the high gamma radiation field was eliminated. The conditions used were:

Pressure	500 psig
Circulation Flow Rate	7.5 gpm
Feed-Bleed Rate	0.1 gpm
Temperature	240°F
Residence Time at Temperature	8 hours

An overall decontamination factor (D.F.*) of 30-60 was obtained. This was the result of a D.F. of 10-15 for the initial fission product-UO_2 decontamination cycle and a D.F. of 3-4 for the final activated corrosion product decontamination cycle.

4.4.1.4.2 S1W Decontamination(10)

In December 1957, the naval reactor prototype (S1W) located at Naval Reactor Facility (NRF), Idaho Falls, Idaho, was decontaminated using a feed-bleed procedure. The purpose of the decontamination was to remove activated corrosion product scale; therefore, a reducing-complexing solution (Table 8) was used at a temperature of 200°F and at a pressure of 300 psig. Some difficulty was encountered during the initial feeding of the solution when the feed pump developed a leak. It was necessary to hold the solution in the makeup tank for several hours during the repair operation. This additional exposure of the solution in the makeup tank probably depleted the solution and resulted in the low D.F. of 1.5-2.0 that was achieved.

4.4.1.4.3 KAPL 120-Loop Decontamination

In April 1957, KAPL 120 loop located at Hanford, Washington, (HAPO) was decontaminated using a fill-flush procedure. (25-26) This loop, unlike WAPD 29, is fabricated of Type 347 stainless steel. Its overall size is larger, having a surface area of 142 ft^2 and a total volume of 75 gallons (40 gallons circulating and 35 gallons in the pressurizer. (27). The loop had been in operation since October 1955. The chief source of activity in the loop was due to the deposition of activated

---

* $\frac{\text{Net Initial Activity}}{\text{Net Final Activity}}$

corrosion products. Consequently, the reducing-complexing solution was used. The in-pile section of the loop was isolated from the rest of the system to avoid exposing the solution to high radiation levels. The conditions used were as follows:

Temperature .... 250°F

Pressure .... 140 psig

Flow Rate .... 30-31 gpm

Residence Time at Temperature. . Approximately 8 hours

An average final D. F. of 2.1 was achieved, measured after the loop had been restored to operating conditions by a series of water flushes.

A second attempt was made to decontaminate the KAPL 120 in May 1957 with the same reducing-complexing solution. In this instance the temperature of the solution was raised for 250°F to 300°F. This second decontamination resulted in an average D. F. of 1.6. The actual dose rates measured in the two decontaminations are given in Table 9.

TABLE 9 Final results of KAPL 120-Loop Decontaminations Using the WAPD Reducing-Complexing Solution (I, II)

	April 1957(I)			May 1957 (II)		
Location of Survey Point	Initial (mr/hr)	Final (mr/hr)	D. F.	Initial (mr/hr)	Final (mr/hr)	D. F.
Pump No. 1 Outlet	320	170	1.9	180	110	1.6
Strainer	450	250	1.8	300	180	1.7
Heat Exchanger	1000	400	2.5	330	290	1.2
Isolation Valve	600	250	2.4	230	180	1.3

The KAPL 120 loop was next decontaminated in July 1958 using the Turco "4501" Process. (28) This process involves the use of three solutions:

Turco 4501 - an alkaline chelating agent containing potassium hydroxide, potassium salts of organic acids, aliphatic amines, phenolics and water.

Turco 4502 - a solid compound used in solution containing approximately 40% potassium hydroxide, 40% potassium carbonate, plus potassium permanganate, inhibitors, and wetting agents.

25% Nitric Acid - passivation treatment.

Preliminary bench scale studies by HAPO personnel gave promising results.(29-31) It was decided to modify the process and substitute 10% citric acid for the 25% nitric acid rinse.

The conditions for the various cycles in the process are tabulated in Table 10.

TABLE 10 Operating Conditions for Modified Turco "4501" Process

Solution	Temp.	Residence Time at Temperature (Minutes)
4501	275°F	60
Water flush	---	
4502 (2 lbs./gal)	220°F	60
Water flush	---	
Citric acid (10% by wgt.)	160°F	30

Continuous water flushes and final treatment with NH_4OH to remove any citric acid remaining in the loop.

The initial decontamination treatment resulted in D. F.s ranging from 1.2 to 20.6 depending upon the location. It was decided to repeat the operation using the procedure as recommended by Turco, i.e. 25% nitric acid rinse supplemented by another treatment with Turco 4501 and the 25% nitric acid. The actual gamma dose rates measured in the two decontaminations are given in Table 11.(32)

Corrosion results from the July 1958 (III) decontamination indicated that the weight losses (mgs/dm^2) on metal specimens ranged from 10.3 for type 304 stainless steel (as annealed) to 21.7 for Type 304 stainless steel (as sensitized), to 590 for Type 410 stainless steel (as annealed). There was no evidence of caustic type corrosion on any of the specimens that were examined. Metallurgical examination of piping, removed after the

TABLE 11 Final Results of KAPL 120 Loop Decontaminations (III, IV)

Turco "4501" Process

Location of Survey Point	July 1958 (III) Turco "4501" Process except for 10% Citric Acid Rinse			August 1958 (IV) Turco "4501" Process Plus Repeat of the Turco 4501 and 25% Nitric Acid Steps		
	Initial (mr/hr)	Final (mr/hr)	D. F.	Initial (mr/hr)	Final (mr/hr)	D. F.
Pump No. 1 Outlet	140	250	---	250	100	2.5
Strainer	2200	230	9.6	230	130	1.8
Heat Exchanger	250	200	1.2	200	70	2.9
Isolation Valve	350	15	20.6	---	---	---

August 1958 (IV) decontamination did not reveal any evidence of intergranular corrosion, stress corrosion, cracking or crack propagation.(32)

A major problem noted with the use of the Turco "4501" Process was the excessive decomposition of the Turco 4502 solution resulting in a large amount of brown maganese dioxide being formed in the system. The process probably depends upon both the loosening action and the chemical reducing power of the Turco 4501 solution. Upon the addition of the Turco 4502, solution, a chemical reaction takes place and the manganese dioxide is formed. This material has a scavenging effect, and much of the dissolved and suspended activity is probably adsorbed on its surface. The removal of this material is then accomplished by peptization (a process which resembles dissolution except the particles are of colloidal size rather than molecular size) with the nitric acid solution. Unless the bulk of the 4501 solution is flushed out of the system, an excessive amount of manganese dioxide will be formed which may not all be dispersed by the nitric acid.

4.4.1.4.4 Oak Ridge Facilities

The decontamination of reactor fuel processing plants at Oak Ridge is achieved through the use of built-in decontaminating equipment.(33) A solution of 30% nitric acid alternated with a solution of 20% sodium hydroxide and 2% sodium tartrate has been found to be successful in decontaminating the "Thorex" and "Purex" plants. A solution of 3% hydrofluoric acid and 20% nitric acid (3-20 reagent) is used when initial treatments prove ineffective. The effectiveness of the latter solution depends to a large extent on the corrosion of the stainless steel surface (34). Corrosion rates of one inch per month have been reported for Type 304L stainless steel in boiling 3-20 reagent (35).

A satisfactory method of removing plutonium and fission product activity in a homogeneous reactor was the use of a chromous sulfate sulfuric acid solution (36). The solution, 0.4 molar $CrSO_4$ - 0.5 molar H_2SO_4, was used at 85°C and with a contact time of 2-4 hours. Stainless steel corrosion rates varied from 200-500 mils/year for a 4.4 hour contact period.

Some care must be employed with the chromous sulfate-sulfuric acid solution as it is easily oxidized by the air. Consequently, an inert atmosphere must be kept over the solution at all times.

4.4.2 Ultrasonic Decontamination Methods

Ultrasonic cleaning has been used effectively to decontaminate small objects. The object to be cleaned is placed in a cleaning tank containing a detergent liquid. A signal generator is then used to produce a sound wave in the liquid. During the rarefaction portion of the sound wave, minute bubbles are formed throughout the bath. During the compression wave these tiny bubbles are compressed until they explode inwardly or implode. The forming and imploding of these bubbles is the phenomenon known as cavitation. In the immediate area of these bubbles tremendous temperatures and pressures exist during the cavitation cycle. The combination of temperatures and pressures, coupled with the force of the implosion, cleans the object. Work performed at Alco Products, Inc. (9) indicated that ultrasonic energy (40 kc/sec) used in conjunction with Oakite No. 131 Cleaner at 80°F was ineffective in removing activated corrosion product scale from samples exposed to APPR-1 primary coolant conditions. However, it was effective in removing activated corrosion product scale from specimens exposed to the same coolant but at a lower temperature and pressure.

Henry (37) showed that the use of ultrasonics enhances the effectiveness of a decontaminating solution. Small samples of contaminated piping were decontaminated at levels of 20-50 kc/sec and at 1000 kc/sec. Some of the conclusions reported are as follows:

1. Ultrasonics are approximately 50% to 80% effective in decontamination.
2. The use of water as a medium has limited effectiveness.
3. The final D. F. of the sample using a water bath can be in-

creased further by continuing ultrasonics with a chemical decontaminating solution.

4. The effectiveness of an ultrasonic system depends on the method of coupling the ultrasonic source relative to the contaminated surface.

5. The use of magnetrostrictos at levels less than 100 kc/sec appears to be most feasible due to size and method of attachment. These low frequency transducers can be clamped directly onto the pipe. Their effectiveness could vary with the distance from the transducer, but it is believed that it may be effective up to 2 feet.

It appears, therefore, that ultrasonics may have some application in decontaminating reactor facilities provided suitable methods can be found to couple the ultrasonic generators to the system. The problem associated with the decontamination of a large heat exchanger, for instance, would indeed be formidable.

4.4.3 Electrolytic Methods

Huff (38) has reported excellent results in decontaminating stainless steel metal surfaces. Surfaces as large as 4 ft.^{2} have been decontaminated using current densities of 100-150 amp/ft^2 with the contaminated item as the anode in solution of either sulfuric or phosphoric acid. Bennett (39) has reported decontamination factors as high as 3000 for Type 347 stainless steel using a similar method.

4.4.4 Other Methods

Various methods have been proposed for decontaminating plants and equipment, some of which have been adapted and used successfully. While most of the adapted methods are excellent for their own particular application, their possible adaptation to the decontamination of a reactor system, in most cases, would require extensive engineering evaluation. Sand blasting has been used effectively by ORNL to decontaminate tank cars (40). Abrasive slurries have also been used with some success. However, it is imperative in these cases that all the abrasive material be removed from the system. Consequently, direct access to the system is a prerequisite to the use of these types of abrasive materials. However, one method which has been proposed for abrasion decontamination is the use of dry ice in pellet form. In this operation it is not necessary to have access to remove the residual material since it will evaporate. The scale removed by the abrasive action of the solidified CO_2 can be subsequently washed out. The possibility of pumping CO_2 pellets and their effectiveness as an abrasive is still unproven.

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4.5 Initial Screening of Chemical Agents Applicable to APPR-1 Decontamination

Phase 2 of the APPR-1 Decontamination Program involved screening a number of chemical agents which, on the basis of available literature and prior experimental work, might prove effective in removing activated corrosion product scale. This was accomplished on a laboratory bench scale (Fig. 7) using samples of APPR-1 primary blowdown piping. These samples (Type 304 Schedule 80) had been exposed to the primary coolant for approximately 7 months. When used for experimentation purposes, these samples had decayed for approximately 4 months. Consequently, activity levels were low enough (1 mr/hr) to permit beta-gamma counting with a thin mica end-window tube connected to a Model 1091 Atomic Associates Scaler.

A sample was initially counted and then placed in a beaker containing the primary chemical solution at a specified temperature. Temperature control was ± 3°C and all solutions were exposed to the atmosphere during the decontamination treatment. Agitation was provided by a mechanical stirrer. After a specified time the sample was removed and recounted. If a secondary rinse solution was used, the sample was placed in a beaker containing this solution. After completion of the treatments the sample was counted again and examined under a microscope for general appearance and evidence of excessive corrosion. A number of samples were submitted for metallographic examination. Comparative results for the different decontamination solutions that were evaluated are given in Table 12. An initial caustic permanganate treatment followed by a nitric acid rinse gave the best results and merited further investigation.

Additional screening results obtained using caustic permanganate and various secondary rinses are tabulated in the Appendix. The experimental procedure was the same as that used to obtain the data in Table 12. Due to the number of variables involved, no special consideration was given to achieving duplicate results; consequently, only general conclusions can be made concerning this data. These are as follows:

1. The removal of the corrosion product scale with its associated activity by the caustic permanganate-rinse method is accomplished primarily by the secondary rinse phase of the treatment. The caustic permanganate treatment removed only about 17% of the total activity removed (D. F. of 1.2).

2. In experiments with 5-10% nitric acid as a secondary rinse, a caustic permanganate solution consisting of 10% sodium hydroxide and 5% potassium permanganate appears to be an optimum concentration (Fig. 8-A).

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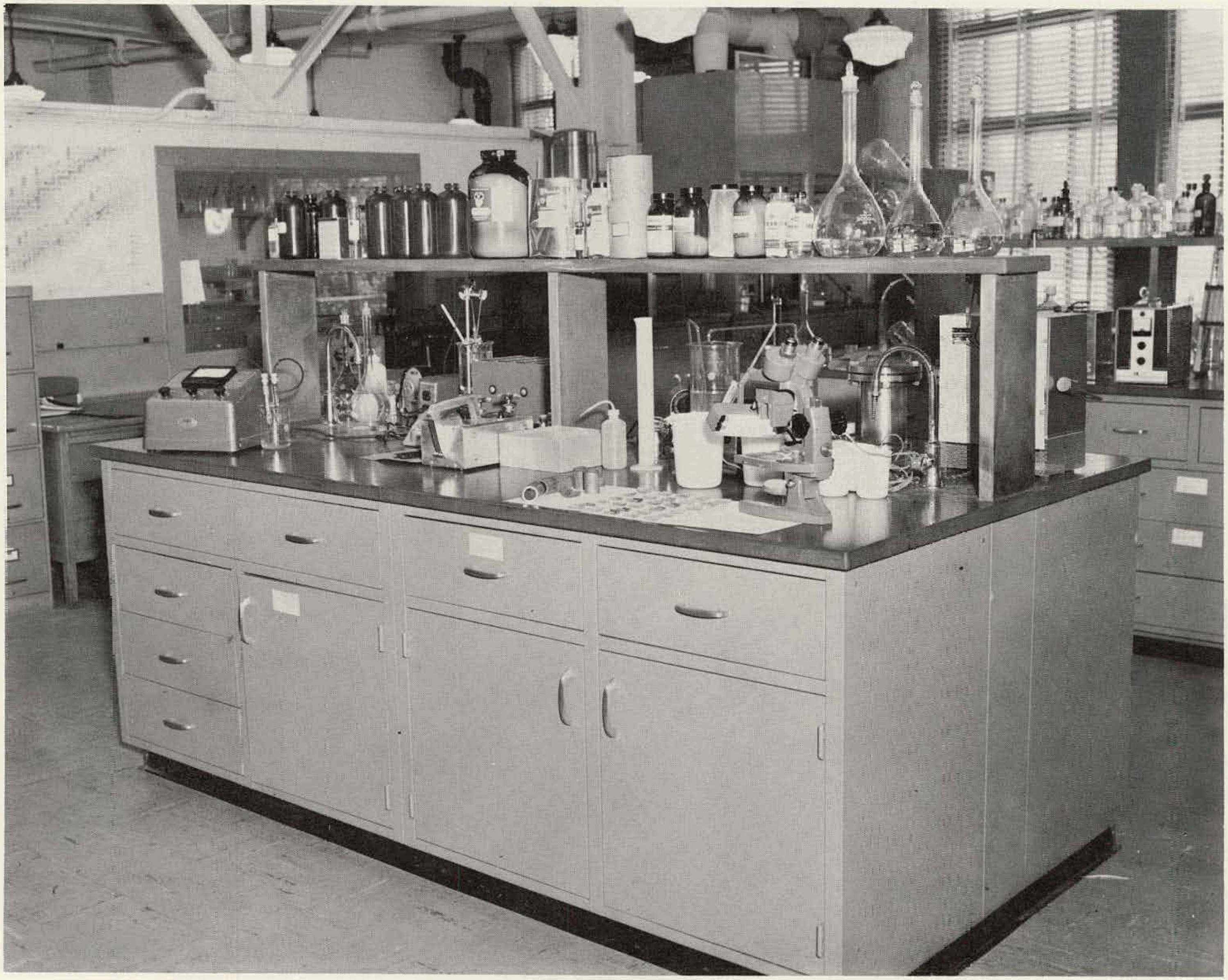

Fig. 7 Laboratory Bench Facilities Used in the Screening of Chemical Decontamination Agents

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TABLE 12 EVALUATION OF VARIOUS DECONTAMINATING SOLUTIONS SCHEDULE 80, TYPE 304SS PIPE FROM APPR-1 PRIMARY BLOWDOWN LINE

Sample No.	Primary Decontamination Solution Description	Conc. by Wgt.	Temp °F	Time Minutes	D. F.*	Secondary Decontamination Solution Description	Conc. by Wgt.	Temp °F	Time Minutes	Total D. F.*
5	"FERLON" Wyandotte Chemical Corp (caustic cleaner)	1 lb/gal	180°	60	<2	None				
6			190°	60	<2	None				
7			200°	60	<2	None				
5	"FERLON" Wyandotte Chemical Corp (caustic cleaner)	2 lb/gal	190°	60	<2	Citric Acid (reagent grade)	5%	185°	30	N.C.+
6			200°	60	<2	Versene Powder (a)	1%	185°	30	N.C.
7			210°	60	<2			185°	30	N.C.
51	"C.R.S." Wyandotte Chemical Corp (caustic cleaner)	10%	195°	60	N.C.	None				
13	Oakite "Rustripper"	2 lb/gal	180°	30	N.C.	Nitric Acid	10% (by volume)	205°	30	<2
			210°	150		Oxalic Acid (reagent grade)	5%	185°	30	<2

*D. F. = Decontamination Factor = $\frac{\text{Initial Activity (net)}}{\text{Final Activity (net)}}$

+N. C. = No change

(a) Versene Powder -
a polyaminocarboxylic acid chelating agent
Dow Chemical Company
Midland, Michigan

TABLE 12 (Continued)

	Primary Decontamination Solution					Secondary Decontamination Solution				
Sample No.	Description	Conc. by Wgt.	Temp °F	Time Minutes	D. F.	Description	Conc. by Wgt.	Temp °F	Time Minutes	Total D. F.
38	Sodium Hydroxide Sodium Gluconate TOTAL	9 parts 1 part 3 lbs/gal	200°	30	1.7	Nitric Acid	10%(vol.)	200°	30	1.4
39	Sodium Hydroxide Sodium Gluconate TOTAL	9 parts 1 part 3 lbs/gal	180°	30	1.2	Nitric Acid	10%	200°	30	1.2
38	Sodium Hydroxide Sodium Gluconate TOTAL	4 parts 1 part 3 lbs/gal	200°	30	N.C.	Nitric Acid	10%	200°	30	1.3
39	Sodium Hydroxide Sodium Gluconate TOTAL	4 parts 1 part 3 lbs/gal	180°	30	N.C.	Nitric Acid	10%	200°	30	1.1
38	Sodium Hydroxide Sodium Gluconate	4 parts 1 part	200°	30	N.C.	Citric Acid + Versene Powder	5% 1/2%	200°	30	1.1
39	Nacconol (wetting agent) TOTAL	1-1/2% 3 lbs/gal	200°	30	N.C.	Ammonium Citrate (reagent grade)	5%	200°	30	1.1

TABLE 12 (Continued)

Sample No.	Primary Decontamination Solution Description	Conc. by Wgt.	Temp °F	Time Minutes	D. F.	Secondary Decontamination Solution Description	Conc. by Wgt.	Temp °F	Time Minutes	Total D. F.
44	Sodium Hydroxide Sodium Tartrate	20% 2%	195°	30	1.1	Nitric Acid	10%	195°	30	1.5
44	Sodium Hydroxide Sodium Tartrate	20% 2%	210°	30	N.C.	Water	-	195°	30	1.3
53	Oxalic Acid	5%	205°	30	1.3	None				
28	Sodium Hydroxide Potassium Permanganate	5% 2.5%	205°	30	N.C.	Nitric Acid	10%	185°	30	10
20	Sodium Hydroxide Potassium Permanganate	5% 2.5%	205°	30	N.C.	Nitric Acid	5%	205°	30	10

3. In experiments with 10% nitric acid and caustic permanganate, a residence time (exposure time) at a given temperature for the secondary rinse beyond 30 minutes did not appreciably increase the decontamination factor (Fig. 8-B).

4. A comparison of the various acid rinses used to remove the activity indicates that all were effective in removing activity. This may indicate that these rinses removed the activity by the same mechanism (Table 13).

5. There was no apparent difference in the effectiveness of the decontamination in the rinse solution temperature range from 185 to 205°F. (Table 13).

6. A chelating agent such as Versene appears to increase the effectiveness of the rinse solution.

TABLE 13 Effect of Different Rinses in Decontamination with Caustic Permanganate At 185° to 205°F for 30 Minutes

X represents a sample tested.

	Secondary Solution Rinse for 30 Minutes								Total Samples Tested
	Nitric Acid		Oxalic Acid		Citric Acid		Ammonium Citrate		
Final Decontamination Factor	185°F	205°F	185°F	205°F	185°F	205°F	185°F	205°F	
Excellent (50 or greater)	X	XXXX	X		X			X	8
Acceptable (20-49)		XX	XX	X		X	X		7
Not Acceptable (less than 20)	XXX		XX					X	6
Total Samples Tested	4	6	5	1	1	1	1	2	21

Visual examination of the samples revealed no evidence of deleterious effects as a result of the caustic permanganate decontamination treatment. There was some evidence of discoloration on several samples, but this was thought to be due to improper washing of the samples after the decontamination

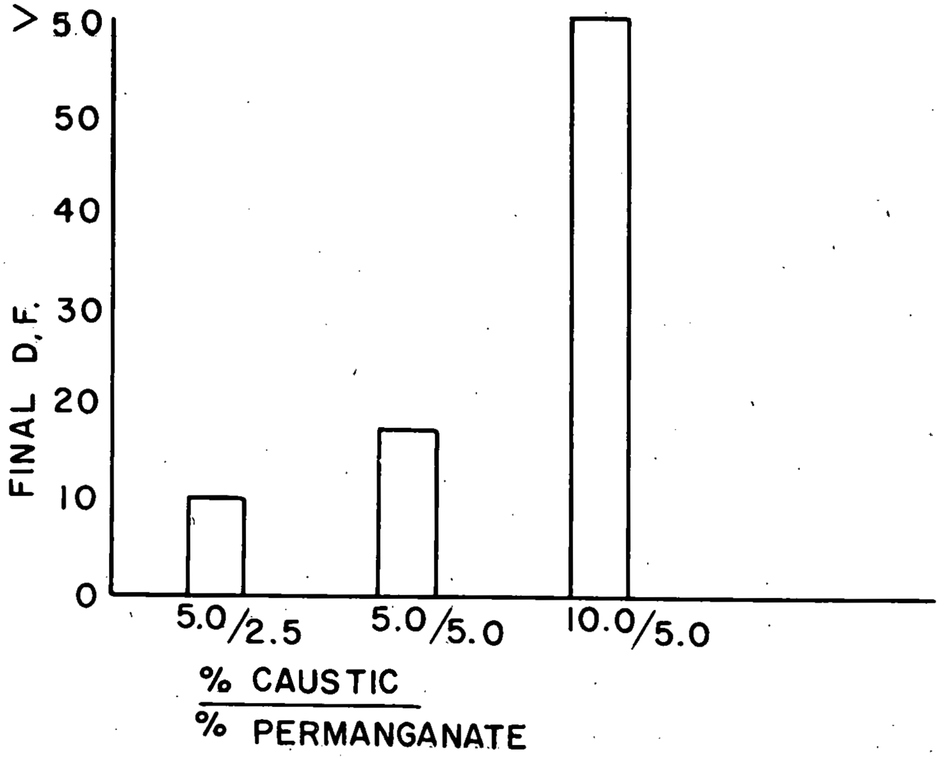

Fig. A

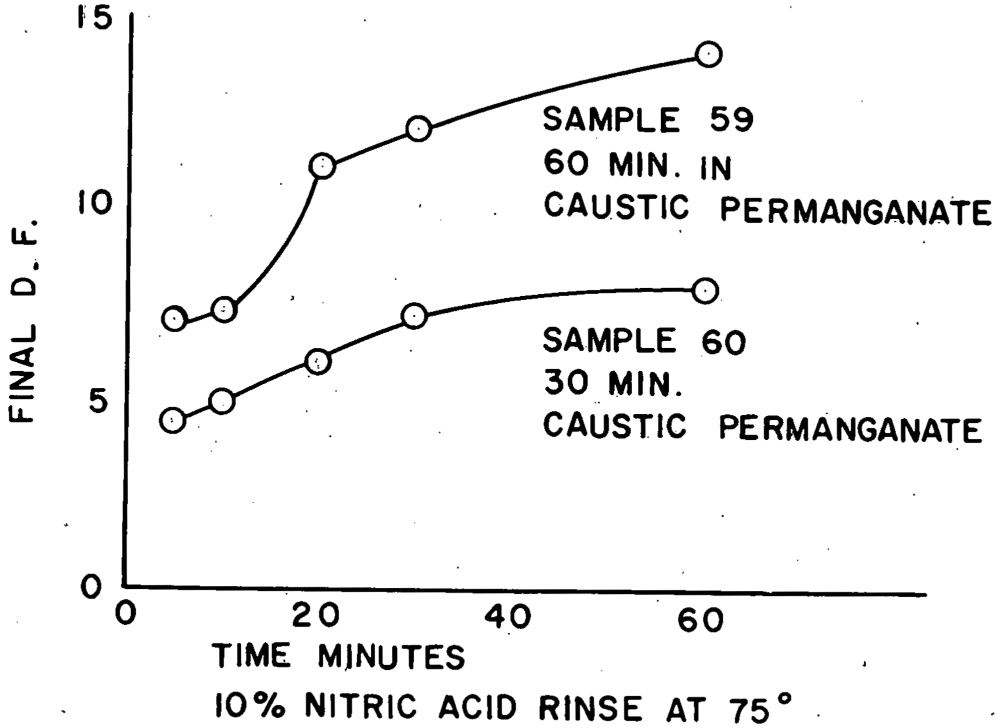

Fig. B

Fig. 8 Results of Initial Screening of Decontamination Solutions

Fig. A Effect of Concentration

Fig. B Effect of Residence Time

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treatment. Metallurgical evaluations of several representative samples were not conclusive. Unfortunately, the original as received samples possessed numerous die marks as a result of the drawing operation during manufacture. (Fig. 9). An attempt to circumvent this limitation by statistical inference using the Poisson Distribution (no. of pits/unit length) did not resolve the problem.

A dynamic autoclave corrosion test was performed to determine the magnitude of corrosion for the caustic permanganate-rinse decontamination treatment (Fig. 1). A 10% oxalic acid rinse was followed by an additional treatment of 10% nitric acid. The oxalic acid would be expected to result in the highest corrosion rate on stainless steel while the nitric acid would be expected to result in the least. The conditions and results of this test are given in Table 14.

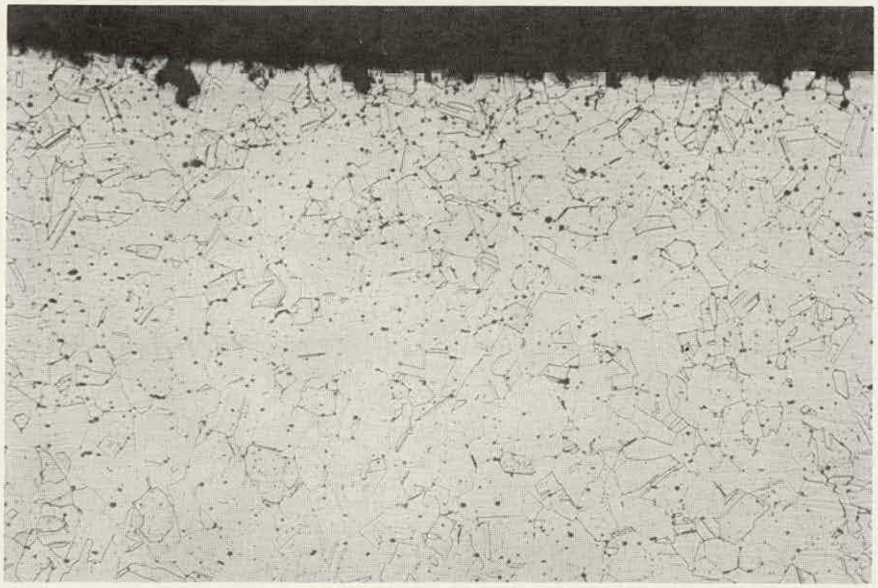
TABLE 14 Dynamic Autoclave Test Results (1 gpm) Caustic Permanganate - Rinse Decontamination Treatment - 1-3/4" x 1/2" x 1/16" Metal Coupons

1. Caustic Permanganate for 60 minutes at 185°F, drain.
2. Demineralized water for 15 minutes at 75°F, drain.
3. Oxalic acid (10%) + 1% Versene Powder for 30 mins. at 175°F, drain.
4. Nitric acid (10%) for 60 minutes at 175°F, drain.
5. Demineralized water for 15 minutes at 75°F, drain.

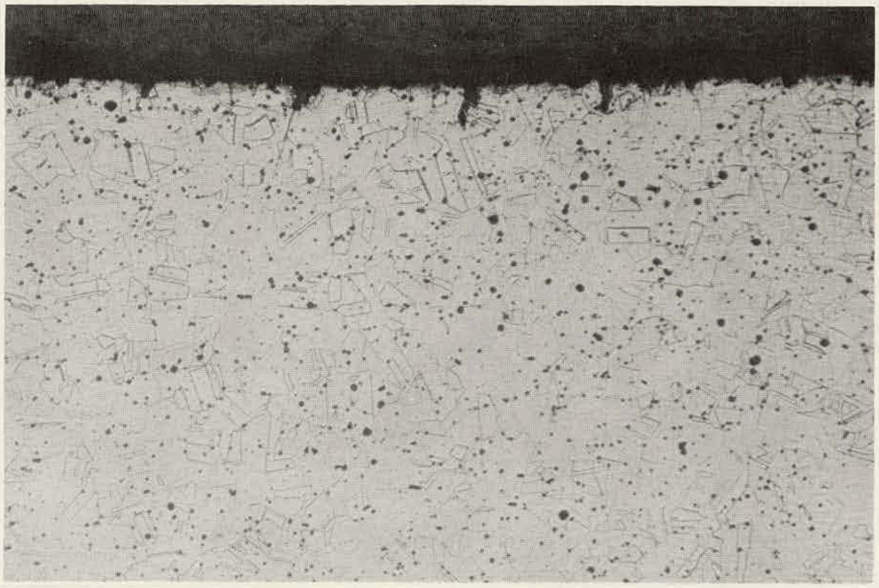
		Weight Loss Results			
		Steps 1-3		Steps 4-5	
Material	Heat Treatment	mgs/dm^2	mils penetration	mgs/dm^2	mils penetration
Type 304 SS	Annealed	18.6	.009	5.4	.003
		20.1	.010	5.9	.003
Type 304 SS	Sensitized 10 mins. at 1200°F, air cool	63.9	.032	13.2	.007
		58.1	.029	15.5	.008

The corrosion results above indicate that the weight loss for steps 1-3 was almost four times as much as the nitric acid and water rinse (steps 4-5). The weight loss results for the sensitized stainless steel was between two and three times as much as the annealed stainless steel. However, it was not felt that the observed results were excessive. Metallurgical examination of the annealed and sensitized stainless steel specimens indicated no evidence of intergranular corrosion or other deleterious effects (Figs. 10, 11).

The three secondary rinses: oxalic acid, citric acid, and ammonium citrate, were evaluated in subsequent loop decontamination studies which form the basis of another report. Nitric acid was not given further consideration as it was felt that the difficulty associated with the storage and handling of this chemical would make decontamination with nitric acid impractical, particularly in a small package type reactor where space economy is paramount.



100 X Oxalic Acid Etch



100 X Oxalic Acid Etch

Fig. 9 Photomicrograph of Type 304 Stainless Steel Pipe

Top:	12846-C	Sample as received
Bottom:	12846-A	Sample exposed to caustic permanganate-ammonium citrate treatment

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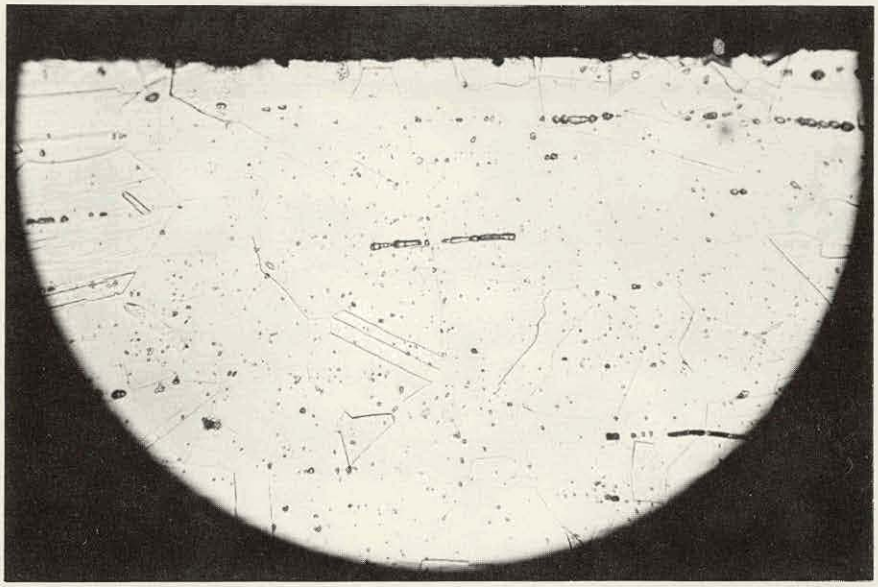
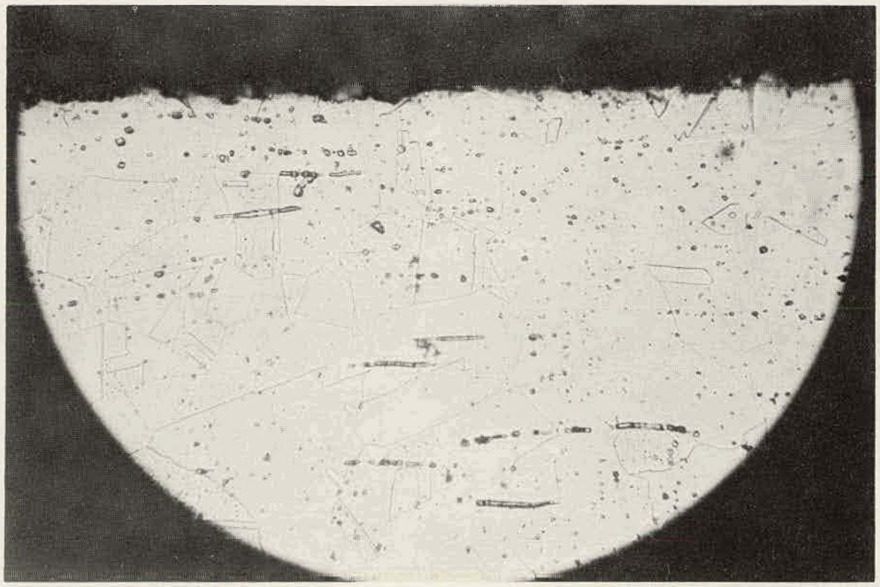


500 X Oxalic Acid Etch



500 X Oxalic Acid Etch

Fig. 10 Photomicrograph of Type 304 Stainless Steel (annealed)

Top: 12847-C Sample untreated

Bottom: 12847-A(C-97) Sample exposed to caustic permanganate-oxalic acid-nitric acid decontamination treatment

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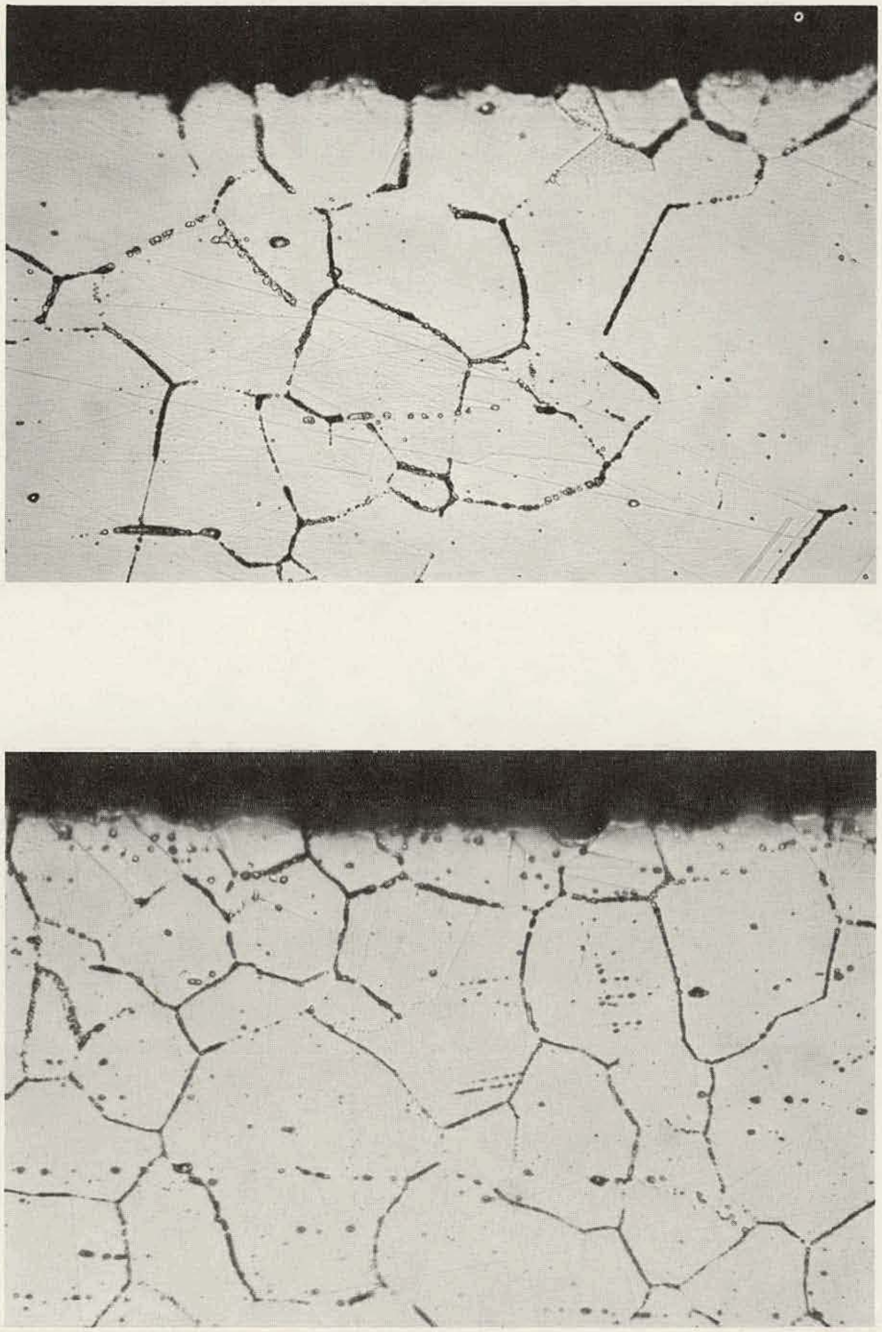

500 X Oxalic Acid Etch

Fig. 11 Photomicrograph of Type 304 Stainless Steel (sensitized)

Top: 12791D Sample untreated

Bottom: 12641B(S-28) Sample exposed to caustic permanganate-oxalic acid-nitric acid decontamination treatment

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APPENDIX

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APPENDIX TABLE I

Evaluation of Caustic Permanganate Decontamination Treatment - 5% Sodium Hydroxide, 5% Potassium Permanganate ("CP55") at 205°F for 30 Minutes; Schedule 80 Type 304 Stainless Steel Pipe Removed from APPR-1 Blowdown Line

		Secondary Decontamination Solution					
Sample No.	"CP55" D. F.	Description	%Conc. by Wgt.	Temp. °F	Time Min.	Total D. F.	REMARKS
40	1.3	Nitric Acid	10%(vol.)	205	30	16	
41	1.4	Nitric Acid	10%(vol.)	205	30	12	30 minute water rinse prior to nitric acid rinse
47	1.1	Nitric Acid	10%(vol.)	205	20	6.3	10 minute water rinse prior to nitric acid rinse
42	1.4	Oxalic Acid	5%	205	2	15	Samples removed and counted at indicated times
					17	27	
					22	31	
					30	35	
43	1.3	Ammonium Citrate	5%	205	2	7.7	Samples removed and counted at indicated times
					10	9.9	
					20	11	
					30	12	
58	1.3	Citric Acid	10%	195	30	11	"CP55" exposure time of 60 minutes

APPENDIX TABLE II

Evaluation of Caustic Permanganate Decontamination Treatment - 10% Sodium Hydroxide, 5% Potassium Permanganate ("CP105") at 185°F for 30 Minutes

Sample No.	"CP105" D. F.	Secondary Decontamination Solution Description	%Conc. by Wgt.	Temp °F	Time Min.	Total D. F.	REMARKS
9	1.3	Nitric Acid	10%(vol.)	185	30	>50	
14	1.2	Nitric Acid	10%	185	30	5.4	No agitation
10, 23	1.2	Nitric Acid	10%	205	30	>50	
34	N.C.	Oxalic Acid	5	185	30	34	
		Versene Powder	1/2				
35	N.C.	"		205	30	17	
30	N.C.	Citric Acid	5	185	30	> 50	
		Versene Powder	1				
36	1.2	1. Citric Acid	5	175	15	3.2	
		2. Nitric Acid	10	175	15	3.8	
59	1.1	Nitric Acid	10	75	5	7.0	"CP105" exposure time of 60 minutes at 195°F
					10	7.3	
					20	11	
					30	12	
					60	14	
60	1.1	Nitric Acid	10	75	5	4.4	"CP105" at 195°F
					10	4.8	
					20	5.8	
					30	7.2	
					60	7.7	

APPENDIX TABLE III

Evaluation of Caustic Permanganate Decontamination Treatment 10% Sodium Hydroxide, 5% Potassium Permanganate ("CP105") at 205°F for 30 Minutes

		Secondary Decontamination Solution					
Sample No.	"CP105" D. F.	Description	%Conc. by Wgt.	Temp. °F	Time Min.	Total D. F.	REMARKS
11, 24	1.6	Nitric Acid	10% (vol)	185	30	17	
8	N. C.	Nitric Acid	5	185	30	15	
12,15,25 29	1.2	Nitric Acid	10	205	30	>50	
46	1.1	Nitric Acid	10	205	20	5.8	10 min. water rinse prior to nitric acid
19	N. C.	Nitric Acid	10	205	10	19	
55	1.1	Oxalic Acid	10	120	30	17	"CP105" exposure time 60 min.
54	1.1	Oxalic Acid	5	75	30	9.1	"CP105" exposure time 60 min.
48, 50	1.2	Oxalic Acid	5	185	15	19	"CP105" exposure time 20 min.
49	1.1	Oxalic Acid	5	185	15	22	"CP105" exposure time 20 min. and 5 minute water rinse prior to oxalic acid
36	-	Oxalic Acid Versene Powder	5 1/2	185	30	>50	
37	-	Versene Powder	1/2	205	30	>50	
16	1.3	Sodium Oxalate	5	205	30	1.3	
21	1.5	Citric Acid Versene Powder	5 1	185	15	15	
26	-	"		185	30	35	Versene did not all dissolve
27	-	"		205	30	49	
32	-	Ammonium Citrate	5	185	30	23	
33	-	"	5	205	30	6	
22	1.9	Citric Acid Versene Powder	5 1	200	30	20	"CP105" at 210°F
17	1.1	Oxalic Acid Versene Powder	5 1/2	205	30	24	"CP105" at 210°F
18	1.1	Ammonium Citrate	5	205	30	> 50	"CP105" at 210°F
22	-	Nitric Acid	10	75	30	5.3	"CP105" at 75°F

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