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REACTORS—POWER

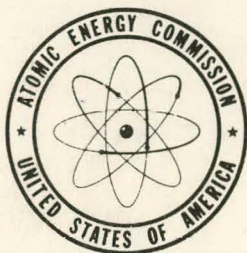
UNITED STATES ATOMIC ENERGY COMMISSION

LITERATURE SURVEY FOR ACTIVITY  
BUILD-UP ON REACTOR PRIMARY  
SYSTEM COMPONENTS

Edited by  
A. L. Medin

January 15, 1958

Alco Products, Inc.  
Schenectady, New York



Technical Information Service Extension, Oak Ridge, Tenn.

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**LITERATURE SURVEY FOR ACTIVITY BUILD-UP  
ON REACTOR PRIMARY SYSTEM COMPONENTS**

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## ABSTRACT

A survey has been made of the possible build-up of long-lived radio-activity on primary system components with increased reactor operation.

Parameters and mechanisms that may contribute to ~~this~~ activity build-up<sup>2</sup> are discussed.

The activity build-up has been attributed to either deposition and/or atom exchange. Experimental data to-date is not sufficient to conclusively attribute the degree to which each of these mechanisms contribute to the build-up. <sup>2</sup>/The need for more experimental information, especially for reactors employing stainless steel clad fuel elements, is discussed.

Theoretical equations to predict the extent of the activity build-up, <sup>and</sup> are discussed. To-date these equations have not been accurate in predicting the build-up of long-lived nuclides.

Decontamination of primary system components may be required to remove this build-up. A discussion of present-day/decontamination agents and their applications<sup>are</sup> is discussed.

**LITERATURE SURVEY FOR ACTIVITY BUILD-UP  
ON REACTOR PRIMARY SYSTEM COMPONENTS**

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## I. INTRODUCTION

Recent information has shown that long-lived nuclide activity tends to build-up activity on primary system surfaces during reactor operation. It is reasonable to assume that this build-up might reach magnitudes which could seriously hinder maintenance of primary system components, e. g., the plugging of a steam generator tube.

Although much effort has been devoted to the solution of this problem, quantitative results are limited. Additional work is necessary before a solution to the problem can be effected. Unfortunately, experimental techniques are necessarily difficult and sampling procedures generally subjective.

Attempts have been made by other installations to theoretically predict the build-up activity. Actual results in most cases have been significantly higher. No comparable results are available for the APPR-1 since it represents the first reactor to employ stainless steel fuel elements.

Alco Products, Inc. under contract with the Army Reactors Branch of the Atomic Energy Commission is now investigating this problem. As part of this program, this report encompasses a survey made of available information on the activity build-up in pressurized water reactors.

### A. COOLANT ACTIVITY

Intrinsic activity <sup>(1)</sup> has been defined as the radioactivity associated with the nuclear composition of the coolant itself. The activation of water theoretically could result in the products shown in Table I.

TABLE-I - IRRADIATION PRODUCTS OF WATER

(n- $\gamma$ )- Products

Parent Nuclide	% Parent Abundance	Product Nuclide	t 1/2	$\gamma$ Energy (Mev)
H <sup>2</sup>	.015	H <sup>3</sup>	12.26 y	None
O <sup>16</sup>	99.759	O <sup>17</sup>	Stable	Stable
O <sup>17</sup>	.037	O <sup>18</sup>	Stable	Stable
O <sup>18</sup>	.204	O <sup>19</sup>	29 s	.19, 1.37, 1.10

(n-p) Products

Parent Nuclide	% Parent Abundance	Product Nuclide	t 1/2	$\gamma$ Energy (Mev)
H <sup>2</sup>	.015	None	None	None
O <sup>16</sup>	99.759	N <sup>16</sup>	7.4 s	6.13, 7.10, 2.7
O <sup>17</sup>	.037	N <sup>17</sup>	4.14 s	n* - 1.0
O <sup>18</sup>	.204	None	None	None

\* n - neutron

The magnitude of the intrinsic activity overshadows the impurity activity, which can be considered composed of all other constituents in the coolant. Generally, the intrinsic activity will be the prime factor in determining the coolant shield, i.e.

$$\frac{\text{intrinsic activity}^{(1)}}{\text{impurity activity}} \approx 300$$

## B. IMPURITY ACTIVITY

Pressurized water reactors now being designed or in operation generally utilize deaerated make-up water. Some naval reactors, however, do not always adhere to this practice. With the addition of non-deaerated water, the presence of certain gases can contribute significantly to the overall impurity water activity. Additional  $N^{16}$  and  $N^{17}$  can form from the dissolved oxygen and nitrogen in the water.

Minor amounts of argon present as an air impurity in the make-up water result in the formation of  $A^{41}$  in the coolant. This nuclide, which has a half-life of 1.82 hours, can contribute significantly to the overall shutdown radioactivity of the primary water. As an example, calculations<sup>(2)</sup> performed for the SAR indicated that the presence of  $A^{41}$  would raise the activity level outside the main primary coolant pumps from 28 to 84 mr/hr measured at fifteen minutes after shut-down.

Since the above gases have short half-lives, radioactive decay will remove their potential hazard. The more important nuclides that contribute to the shut-down activity arise from corrosion products. Considered of minor importance are impurities present in the make-up water. Table II

illustrates some impurities which may be present in the make-up water.

TABLE - II NUCLEAR CHARACTERISTICS OF  
SOME COMMON WATER IMPURITIES

Product Nuclide	% Parent Abundance	Nuclear Reaction	Cross Section (barns)	Product t 1/2	$\gamma$ Energies (Mev)
Na <sup>24</sup>	100	Na <sup>23</sup> (n, $\gamma$ )Na <sup>24</sup>	0.53	15.0h	2.75; 1.37
Ne <sup>23</sup>	100	Na <sup>23</sup> (n, p)Ne <sup>23</sup>	----	40 s	0.44; 1.65
Na <sup>25</sup>	10.1	Mg <sup>25</sup> (n, p)Na <sup>25</sup>	----	60 s	0.98; 0.58; 0.38
Mg <sup>27</sup>	11.1	Mg <sup>26</sup> (n, $\gamma$ )Mg <sup>27</sup>	0.03	9.5m	0.84; 1.02; 0.18
P <sup>32</sup>	100	P <sup>31</sup> (n, $\gamma$ )P <sup>32</sup>	0.20	14.5d	None
Si <sup>31</sup>	100	P <sup>31</sup> (n, p)Si <sup>31</sup>	----	2.62h	1.26
P <sup>33</sup>	0.75	S <sup>33</sup> (n, p)P <sup>33</sup>	----	25 d	None
S <sup>35</sup>	4.2	S <sup>34</sup> (n, $\gamma$ )S <sup>35</sup>	0.26	87 d	None
P <sup>34</sup>	4.2	S <sup>34</sup> (n, p)P <sup>34</sup>	----	12.4s	2.1; 4.0
Cl <sup>36</sup>	75.5	Cl <sup>35</sup> (n, $\gamma$ )Cl <sup>36</sup>	4.4	3.1x10 <sup>5</sup> y	None
Cl <sup>38</sup>	24.47	Cl <sup>37</sup> (n, $\gamma$ )Cl <sup>38</sup>	0.56	37.3 m	2.1; 1.6
Ca <sup>41</sup>	96.9	Ca <sup>40</sup> (n, $\gamma$ )Ca <sup>41</sup>	0.2	1.1x10 <sup>5</sup> y	None
K <sup>42</sup>	0.64	Ca <sup>42</sup> (n, p)K <sup>42</sup>	----	12.5 h	1.5; 0.32

### C. PREDICTED PRIMARY SYSTEM COMPONENT ACTIVITY

Theoretical calculations have been employed to predict activity levels both for the coolant and the impurities in the coolant. In the Naval Reactor Program, impurity activity level calculations were made for various intervals of full power hours operation<sup>(3)</sup>. Table III indicates the results obtained on the measured activity levels versus the predicted levels. As will be indicated later,  $\text{Co}^{60}$  is the major contributor to the activity build-up on the primary components due to its long half-life and energetic gammas. This value was found to be much higher than predicted.

TABLE III OBSERVED VERSUS PREDICTED  
SPECIFIC ACTIVITY  
(S-1-W DATA AFTER 93 FULL POWER HOURS)

Nuclide	Measured ( $\mu\text{c/cc}$ )	Predicted ( $\mu\text{c/cc}$ )
$\text{Mn}^{56}$	$1.9 \times 10^{-1}$	$1.75 \times 10^{-1}$
$\text{Co}^{60}$	$5.8 \times 10^{-5}$	$9.02 \times 10^{-7}$
$\text{Fe}^{55}$	$9.1 \times 10^{-5}$	$5.43 \times 10^{-2}$
$\text{Fe}^{59}$	$1.6 \times 10^{-5}$	$6.37 \times 10^{-6}$
$\text{Cr}^{51}$	$3.8 \times 10^{-4}$	$3.37 \times 10^{-5}$

This difficulty in predicting actual specific activities has been encountered in practically all other pressurized water programs, including the APPR. This inability to make accurate predictions emphasizes the necessity of a combined experimental and theoretical program.

Radiation emanating from primary components after shut-down have been measured at both the S-1-W and APPR. Table IV shows external radiation levels on the S-1-W heat exchanger.<sup>(4)</sup>

TABLE IV. EXTERNAL RADIATION LEVELS AT  
S-1-W HEAT EXCHANGER

	Initial Level (mr/hr)	Level During Full Power (mr/hr)	Level at 10% Power (mr/hr)
Stainless steel boiler	160	600	350 - 400
Carbon steel boiler	0	400	250 - 300

From these, plus additional reactor data and loop tests, investigators became cognizant of the potential serious build-up of radioactivity on system components. Today, most reactor designers are becoming aware of the complicated maintenance problems that may arise from this activity build-up.

## II. ACTIVATION FROM MATERIALS OF CONSTRUCTION

### A. MECHANISM OF IMPURITY ACTIVATION

Since the half-life of the intrinsic coolant activity is small, its contribution to the after shut-down level is rapidly eliminated. The coolant impurities, however, have both long and short half-lives. The longer half-life impurity nuclides are the major contributors to the after shut-down levels.

The principal mechanisms by which impurities become activated and can normally enter the primary coolant, are as follows:

1. Solids or gases which are present in the make-up water and become activated.
2. Recoil of activated nuclides from the core surface which enter coolant.
3. Fuel element materials and other in-flux structural materials which become activated and become released to the coolant.
4. Structural materials which are released from out of flux regions and become activated during residence in flux areas.

The first mechanism, which was discussed previously, does not normally contribute significantly to the activity build-up. (Reference to activity build-up, henceforth in this report refers to the after shut-down levels remaining on primary system components). Most reactor installations, under design or in operation, have utilized demineralized water for make-up to the primary system; thus the impurity addition is negligible. With the exception of  $A^{41}$  which results from the use of non-deaerated water, make-up water impurity

activation is of no significance to the activity build-up problem.

Recoil, as a possible major source of activity build-up, is not considered probable. A uniform corrosion deposit usually forms on the cladding of fuel elements. Since the general range of recoil atoms is in the order of  $10^{-5}$  cm and the deposit thickness is usually of greater magnitude, the number of nuclides that will enter the coolant by this mechanism is considered negligible. Recoil contribution calculations can be shown to have only minor effect on the activity build-up.

Mechanisms 3 and/or 4 are the major contributors to the activity build-up of activity on the primary system. Factors of importance pertaining to these two mechanisms are discussed below.

#### B. FUEL ELEMENT STRUCTURAL MATERIAL

Erosion and corrosion of activated fuel element cladding material can be a major contributor to the activity build-up. Naval Reactors generally employ alloys of zirconium while the APPR-1 uses Type 304 stainless steel for fuel element claddings. Correlation of experimental activity build-up results of the two programs should not be expected for this reason. Table V shows the nominal chemical composition of zirconium alloys<sup>(5)</sup> and Table VI shows Type 304 stainless steel.<sup>(6)</sup>



TABLE V - NOMINAL COMPOSITION OF ZIRCALOYS

Element	Zircaloy-1 (percent)	Zircaloy-2 (percent)	Zircaloy-3 (percent)
Sn	2.2 - 2.8	1.3 - 1.6 0.20	0.20 - 0.30
Fe	0.04- 0.08	0.09- 0.14	0.20 - 0.30
Cr	0.015 max.	0.07- 0.11	0.05
Ni	0.005 max.	0.04- 0.06	0.05
N	0.006 max.	0.006 max.	0.01 max.
Bal.	Sponge Zr	Sponge Zr	Sponge Zr

TABLE VI - NOMINAL COMPOSITION TYPE 304 STAINLESS STEEL

Element	Percent
C (max)	.08
Mn (max)	2.0
Si (max)	1.0
Cr	10.0 - 20.0
Ni	8.0 - 11.0
S (max)	.03
P (max)	.045

Cobalt content in stainless steel is of major importance to the activity build-up. In commercial grade stainless steel, the cobalt content will range as high as 0.2%. Since it is of no significance to the metallurgical or mechanical characteristics of the steel, it is seldom reported, though considered

a minor impurity. Recently, specifications have been issued by certain reactor designers designating very low cobalt stainless steel. To produce this type of steel requires special batch manufacture with resulting increase in steel prices.

Alloys and impurities in other grades of stainless steel and zirconium alloys are of significance to the activity build-up. Included among these alloys are tantalum and niobium. Maximum impurity contents for zirconium alloys are shown in Table VII<sup>(5)</sup>:

**TABLE VII - MAXIMUM IMPURITY CONTENT OF ZIRCALOYS**

Element	Zircaloy-2 (ppm)	Zircaloy-3 (ppm)
Aluminum	75	75
Boron	0.5	0.5
Cadmium	0.5	0.5
Carbon	500	500
Cobalt	20	20
Copper	50	50
Hafnium	200	200
Hydrogen	50	50
Lead	130	130
Magnesium	20	20
Manganese	50	50
Molybdenum	50	50
Oxygen	2500	2500
Silicon	100	100
Titanium	50	50
Tungsten	100	100
Vanadium	50	50

### C. EXPECTED NUCLIDES FROM IN-FLUX MATERIALS

The activation of fuel element cladding and other in-flux material and the subsequent release of these materials to the coolant are major contributors to the activity build-up. While these impurities, even those with relatively long half-lives and with penetrating gamma rays, do not generally contribute

significantly to the overall vapor container shielding requirements, their presence seriously complex the maintenance problem.

Some of the possible nuclides which can result from the use of the zirconium alloys are listed in Table VIII. Of particular interest, is that only  $\text{Zr}^{95}$  has been reported as having been present in any measurable quantity either in the coolant or in the surface deposits.

Table IX indicates possible nuclides which could be formed from Type 304 stainless steel. Of those listed  $\text{Fe}^{59}$ ,  $\text{Mn}^{54}$ ,  $\text{Cr}^{51}$ ,  $\text{Co}^{58}$ ,  $\text{Co}^{60}$ ,  $\text{Mn}^{56}$  and/or  $\text{Ni}^{65}$  have been identified in the APPR-1 primary water.

TABLE VIII - POSSIBLE NUCLIDES FORMED FROM ELEMENTS  
OF ZIRCALOY-2

Parent Nuclide	Parent % Abundance	(n- $\gamma$ Products)			(n-p Products)		
		Product Nuclide	Product t 1/2	Product $\gamma$ Radiations (Mev)	Product Nuclide	Product t 1/2	Product $\gamma$ Radiations (Mev)
Zr <sup>90</sup>	51.5	Zr <sup>91</sup>	stable	stable	Y <sup>90</sup>	64.0 h	none
Zr <sup>91</sup>	11.2	Zr <sup>92</sup>	stable	stable	Y <sup>91</sup>	58 d	1.19
Zr <sup>92</sup>	17.1	Zr <sup>93</sup>	9x10 <sup>5</sup> y	.029	Y <sup>92</sup>	3.5 h	0.94, 0.21
Zr <sup>94</sup>	17.4	Zr <sup>95</sup>	65 d	0.75, 0.72, 0.23	Y <sup>94</sup>	17 m	1.4
Zr <sup>96</sup>	2.8	Zr <sup>97</sup>	17 h	0.75, 0.66	none	----	----
Sn <sup>112</sup>	1.02	Sn <sup>113</sup>	112 d	0.26	In <sup>112</sup>	21 m	
Sn <sup>114</sup>	0.69	Sn <sup>115</sup>	stable	stable	In <sup>114</sup>	14 m	
Sn <sup>115</sup>	0.38	Sn <sup>116</sup>	stable	stable	In <sup>114</sup>	49 d	0.72, 0.55,
Sn <sup>116</sup>	14.3	Sn <sup>117</sup>	14 d	0.162	In <sup>115</sup>	72 s	1.28
Sn <sup>117</sup>	7.6	Sn <sup>118</sup>	stable	stable	In <sup>115</sup>	4.5 h	none
Sn <sup>118</sup>	24.1	Sn <sup>119</sup>	275 d	0.024	In <sup>116</sup>	54.0 m	1.27, 1.09,
Sn <sup>119</sup>	8.5	Sn <sup>120</sup>	stable	stable	In <sup>116</sup>	13 s	2.1, 1.5
Sn <sup>120</sup>	32.5	Sn <sup>121</sup>	1 y	none	In <sup>117</sup>	1.9 h	.16, .56,
Sn <sup>122</sup>	4.8	Sn <sup>123</sup>	27 h	1.5	In <sup>117</sup>	1.1 h	.16
Sn <sup>124</sup>	6.1	Sn <sup>125</sup>	130 d	0.33, 1.4	In <sup>118</sup>	4.5 m	
			40 m	0.20	In <sup>118</sup>	1 m	none
			9.5		In <sup>119</sup>	18 m	none
			10 d		none	none	none
					none	none	none
					none	none	none
					none	none	none

TABLE IX - POSSIBLE NUCLIDES FROM ELEMENTS  
OF TYPE 304 STAINLESS STEEL

Parent Nuclide	% Parent Abundance	<u>(n-<math>\gamma</math>) Products</u>			<u>n-p Products</u>		
		Product Nuclide	t 1/2	Gamma Radiation (Mev)	Product Nuclide	t 1/2	Gamma Radiation (Mev)
Fe <sup>54</sup>	5.9	Fe <sup>55</sup>	2.9 y	none	Mn <sup>54</sup>	2 m 300 d	0.84
Fe <sup>56</sup>	91.6	Fe <sup>57</sup>	stable	stable	Mn <sup>56</sup>	2.58 h	0.85, 1.8, 2.1, 3.
Fe <sup>57</sup>	2.2	Fe <sup>58</sup>	stable	stable	Mn <sup>57</sup>	1.7 m	0.12, 0.13, 0.7
Fe <sup>58</sup>	0.33	Fe <sup>59</sup>	45 d	1.10, 1.29, 0.19	none	none	none
Cr <sup>50</sup>	4.4	Cr <sup>51</sup>	27 d	0.32	V <sup>50</sup>	stable	stable
Cr <sup>52</sup>	83.7	Cr <sup>53</sup>	stable	stable	V <sup>52</sup>	3.77 m	1.4
Cr <sup>53</sup>	9.5	Cr <sup>54</sup>	stable	stable	V <sup>53</sup>	2.0 m	1.0
Cr <sup>54</sup>	2.4	Cr <sup>55</sup>	3.6 m	none	V <sup>54</sup>	55 s	0.99, 0.84 2.2
Ni <sup>58</sup>	68.0	Ni <sup>59</sup>	8x10 <sup>4</sup> y	none	Co <sup>58</sup>	71 d, 9 h	0.81, 1.64
Ni <sup>60</sup>	26.2	Ni <sup>61</sup>	stable	stable	Co <sup>60</sup>	5.2y, 10.5m	1.33, 1.17
Ni <sup>61</sup>	1.1	Ni <sup>62</sup>	stable	stable	Co <sup>61</sup>	1.65 h	.07
Ni <sup>62</sup>	3.7	Ni <sup>63</sup>	80 y	none	Co <sup>62</sup>	14 m, 1.6 m	1.2, 1.0
Ni <sup>64</sup>	1.0	Ni <sup>65</sup>	2.56 h	1.5, 1.12, 0.37	none	none	none
Mn <sup>55</sup>	100	Mn <sup>56</sup>	2.58 h	0.85, 1.8, 2.1, 3	Cr <sup>55</sup>	3.6 m	none

#### D. NUCLIDES FROM STRUCTURAL MATERIALS

To date, austenitic type stainless steel is the major material of construction employed for reactor primary systems. Minor amounts of Stellite, Haynes 25, and 17-4 PH steel are also employed throughout the primary system in Naval Reactors and the APPR. A carbon steel boiler was also employed at the S-1-W reactor. Typical compositions<sup>(8)</sup> of these materials of construction are listed in Table X.

The so-called minor impurities which may be present in any of the materials of construction have been found to be important. Reference to the previous statements on the cobalt impurity emphasizes this point.

TABLE X - STRUCTURAL MATERIAL COMPOSITIONS

Element	A-212 B (Percent)	Haynes 25 (Percent)	Armco 17-4 PH (Percent)	Stellite 19 (Percent)
C	0.35	0.15 max	.07 max	1.8
Mn	0.90	1.0 - 2.0	1.0 max	
P	0.04		0.04 max	
S	0.04		0.03 max	
Si	0.15-0.30	1.0 max	1.0 max	
Ni		9.0-11.0	3.5-5.0	
Cb-Ta			0.25-0.45	
Cr		19.0-21.0	15.5-17.5	31
Cu			3.5- 4.5	
Fe	Bal.	2.0 max	Bal.	3
W		14.0-16.0		10
Co		Bal.		52
Others				2

#### E. CORROSION RATES

Reporting of corrosion rates is based on either material weight loss or weight gain. From the standpoint of the activity build-up, the corrosion release rate is of prime importance. Unless material enters the coolant, it can

not be redeposited on primary system components. As an example, austenitic Type 304 stainless steel has been reported as having corrosion rates of approximately 5 to 10 mg/dm<sup>2</sup>/mo at water conditions normally found in a primary system. However, the release rate of this material has been estimated as low as 2 to 3 mg/dm<sup>2</sup>/mo.

The literature seldom differentiates between corrosion rate and release rate. In Table XI, corrosion rates <sup>(8, 9)</sup>(not release rates) are given for typical materials of construction for primary system structural materials or cladding. Since the release rates are not generally reported, one assumes the worse case in calculations, that being the release rate is equal to the corrosion rate.

#### F. CRUD COMPOSITION

The insoluble corrosion release products tend to conglomerate as a magnetic mass and have been referred to as crud. The actual experimental data available on the chemical composition of this primary system crud is inconsistent. Iron compositions in the crud have varied from as low as 20% to as high as 90%. Examples of typical crud experimental values <sup>(10)</sup> are shown in Table XII and XIII. The data is basically on reactors employing zirconium alloys as fuel element cladding materials. For this reason, correlation between this data and that which may be expected from the APPR-1 is again difficult. It is interesting to note that the compositions show little or no zirconium chemically.

**TABLE XI - CORROSION RATES OF COMMON PRIMARY SYSTEM MATERIALS  
EXPOSED TO HIGH TEMPERATURE, HIGH PURITY WATER**

Material	Temp. (°F)	Velocity (f. p. s)	O <sub>2</sub> (cc/kg)	H <sub>2</sub> (cc/kg)	pH	Descaled Corr. Rate (mg 1dm <sup>2</sup> -mo.)
T 304 SS	500	30	1-5	N. A.	7	5
T 304 SS(ELC)	500	30	D.	25-50	7	5
T 347 SS	500	30	1-5	N. A.	7	5
A-212 B	600	30	0.1	D.	11	30
A-302	600	2	0.10	20	9.5	72
Zr-2	500	10-20	D.	100	8.5	5
Hf	500	20	30	100	7	5
Haynes-25	500	30	N. A.	100	7	5
T 410 SS	500	1/60, 30	1-5	N. A.	7-10	5
T 440 C SS	500	30	D	N. A.	7	15

Symbols: D  $\bar{x}$  degassed water, less than 0.5 cc of oxygen (STP) per kilogram of water

NA - no analysis made for constituent. When NA appears in hydrogen column, the implication is that hydrogen is probably present in the range of 0 to 20 cc of hydrogen (STP) per kilogram of water



**TABLE XII. CHEMICAL COMPOSITION OF CRUD IN A DEGASSED SYSTEM AT A FLOW RATE OF 6 FEET PER SECOND**

Element	Wet Chem. (% Crud)	Spect. (% Crud)
Fe	86.2	73.5
Ni	6.45	10.5
Cr	1.75	2.9

**TABLE XIII. CHEMICAL COMPOSITION OF CRUD IN A HYDROGENATED SYSTEM**

Element	Flow Rate, 6 ft/sec		Flow Rate, 27 ft/sec	
	Wet Chem. (% crud)	Spect. (% crud)	Spect. % crud	Spect % crud
Fe	60.0	71.0	74.9	80.6
Ni	6.68	15.0	6.27	2.98
Cr	3.11	5.4	13.0	7.25

### III. OPERATING REACTOR DATA

#### A. OBSERVED NUCLIDES IN COOLANT

Data have been collected on nuclides experimentally found to be present in the coolant of the S-1-W and the APPR-1. For this report, results obtained through APPR-1 700 hour test are given. Data from the S-1-W and the APPR-1 can not be directly correlated due to difference in the materials of construction, core cladding and other physical and chemical characteristics.

Table XIV illustrates the percentage of long-lived nuclides found in the crud of the S-1-W<sup>(11)</sup> and the coolant of the APPR.<sup>(7)</sup> Since the analyses are on a different basis, i. e., crud versus coolant impurity activity, the results can not be quantitatively compared. Of particular interest is the larger total cobalt percentage found in the APPR-1 system. This may be partially due to the difference in assay techniques.

Manganese activity was not found in the S-1-W crud but only in solution and, therefore, it is assumed all corrosion products of manganese are in solution. Since it is soluble, manganese is frequently used in calculating release rates. By assuming that the corrosion products have same chemical percentage as the base metal, determination of the manganese activity can be correlated with the release rates. This assumption has not been found to be completely valid.

In the S-1-W, the large hafnium activity results from abrasion of the hafnium control rod rubbing shoes. Hafnium is not used at the APPR-1 and this activity along with the  $\text{Zr}^{95}$  activity would not, of course, be present.

TABLE XIV OBSERVED NUCLIDES

Nuclide	S-1-W * % of Total	APPR-1 ** % of Total
Co <sup>60</sup>	19.5	55.4
Co <sup>58</sup>	5.5	
Cr <sup>51</sup>	1.1	0.4
Zr <sup>95</sup>	0.7	----
Fe <sup>59</sup>	0.7	7.3
Hf <sup>181</sup> (175)	53.3	----
Mn <sup>54</sup>	----	14.2
Total	80.8	77.3

\* insoluble crud (average of nine values)

\*\* circulating water and crud (average of six values)

At both the S-1-W and the APPR-1, the major contributors to the total long-lived activity results from only a few nuclides. These nuclides result from impurities in the materials of construction and again illustrate their importance.

At the end of the S-1-W first core, the plant was shut down, the water drained and a carbon steel boiler was installed as the port boiler for experimental purposes. At this shut-down, significant deposits of activity were observed in the starboard stainless steel boiler. An external dose rate of 300 mr/hr was measured.<sup>(12)</sup> Nuclides identified in the deposited material included Co<sup>60</sup>, Co<sup>58</sup>, Fe<sup>59</sup>, Hf<sup>181</sup> and Ta<sup>182</sup>.

## B. APPR-1 SURVEY DATA

Deposited activity was observed at the APPR-1 after the 700 hour full power test. Survey data from the first shutdown and one subsequent shutdown are presented in Table XV<sup>(13)</sup>.

TABLE XV SURVEY DATA FROM APPR-1

Location of Survey Point	Dose Rate (mr/hr)		Decay Data			
	First *	Second **	First Shutdown % of Total	t 1/2	Second Shutdown % of Total	t 1/2
Flow Tube Taps	150	110	62 38	170 hr 7 hr	60 40	3 hr long
Side of Steam Generator	45	24	50 50	87 hr 3 hr	46 54	245 hr 5 hr
Pressurizer Elbow	310	110	1 component long-lived		1 component long-lived	

\* Corrected to 30 hours after shutdown. First measurement 32 hours after shutdown.

\*\* Corrected to time of shutdown. First measurement 5.67 hours after shutdown.

For both shutdowns, primary water was present in the system when the survey was made. After the first shutdown, which occurred at the end of the 700 hour test, the system was drained to demonstrate (as part of the contract) that a tube in the steam generator could be plugged. When the head of the steam generator was dropped the dose rate from the deposited material was measured at levels of 250-300 mr/hr.<sup>(13)</sup> The same approximate dose rate was measured emanating from the tube bundle.

Some tentative observations for this APPR data are given below:

1. A quantitative and qualitative difference existed for the activity deposited throughout the system. These differences are shown by the decay data shown in Figure 1 and the dose rate readings in different parts of the system.
2. The deposited activity is partly composed of a loosely bound film and an adherent film as noted by the decrease in dose rate level after the second shutdown. Some of the loose bound film must have been removed in draining the system during the shutdown.
3. The remaining deposited film evidently is sufficiently adherent to remain after draining the system.

#### C. DEPOSITED ACTIVITY

Radiochemical data obtained<sup>(14)</sup> during the period from May 28, 1956 to August 11, 1956 at the S-1-W are summarized in Table XVI. For the results shown, three values were obtained of the cpm/mg for the SFR edge bonded sub-assembly wipe sample of each nuclide. The small amount of crud collected was not weighable and as such did not allow direct specific activity determinations.

FIG.1

DECAY DATA-APPR-I SECOND SHUTDOWN

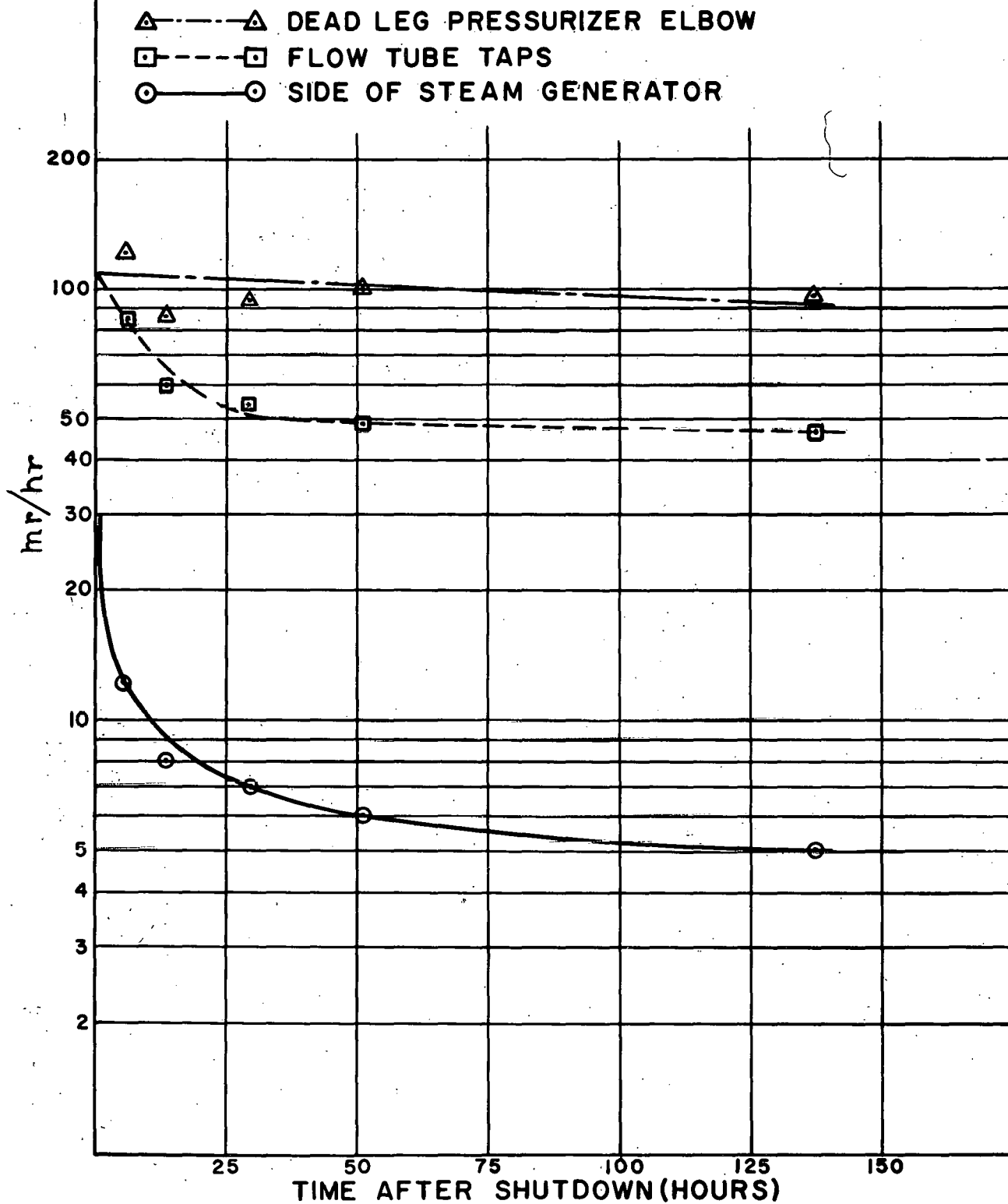


TABLE XVI. WIPE SAMPLES FOR SFR EDGE BONDED SUBASSEMBLY

Nuclide	Activity (cpm/mg)	Nuclide	Activity (cpm/mg)
$\text{Co}^{60}$	$8.35 \times 10^6$	$\text{Zr}^{95}$	$4.31 \times 10^5$
	$8.20 \times 10^6$		$4.24 \times 10^5$
	$3.96 \times 10^6$		$2.04 \times 10^5$
$\text{Co}^{58}$	$2.04 \times 10^6$	$\text{Fe}^{59}$	$2.78 \times 10^5$
	$2.00 \times 10^6$		$2.74 \times 10^5$
	$9.66 \times 10^5$		$1.32 \times 10^5$
$\text{Cr}^{51}$	$4.35 \times 10^5$	$\text{Hf}^{181} (175)$	$5.23 \times 10^6$
	$4.27 \times 10^5$		$5.14 \times 10^6$
	$2.06 \times 10^5$		$2.48 \times 10^6$

Wipe samples can generally only show the activity of the loose deposits. It is conceivable that the adherent film might have different composition than this wipe sample although experimental data is lacking to demonstrate this point.

Table XVII shows results<sup>(15)</sup> from this same operating period on water and crud. Of interest is the ratio of crud activity to the total water and crud activity. Of significance is the wide variation of the percentage of crud activity in the water, ranging from 25 to 100%. Should the former value be correct, it would appear that the purification system sees more soluble nuclides than insoluble.

TABLE XVII RATIO OF CRUD ACTIVITY TO TOTAL ACTIVITY

Total Activity (cpm/ml)	Crud Activity (cpm/ml)	Ratio of Crud to Total Activity
2180	748	0.34
2500	756	0.30
1990	504	0.25
1410	495	0.35
1850	675	0.36
2142	760	0.35
1820	878	0.48
950	960	1.01
1150	625	0.54
1210	863	0.71
1440	950	0.66

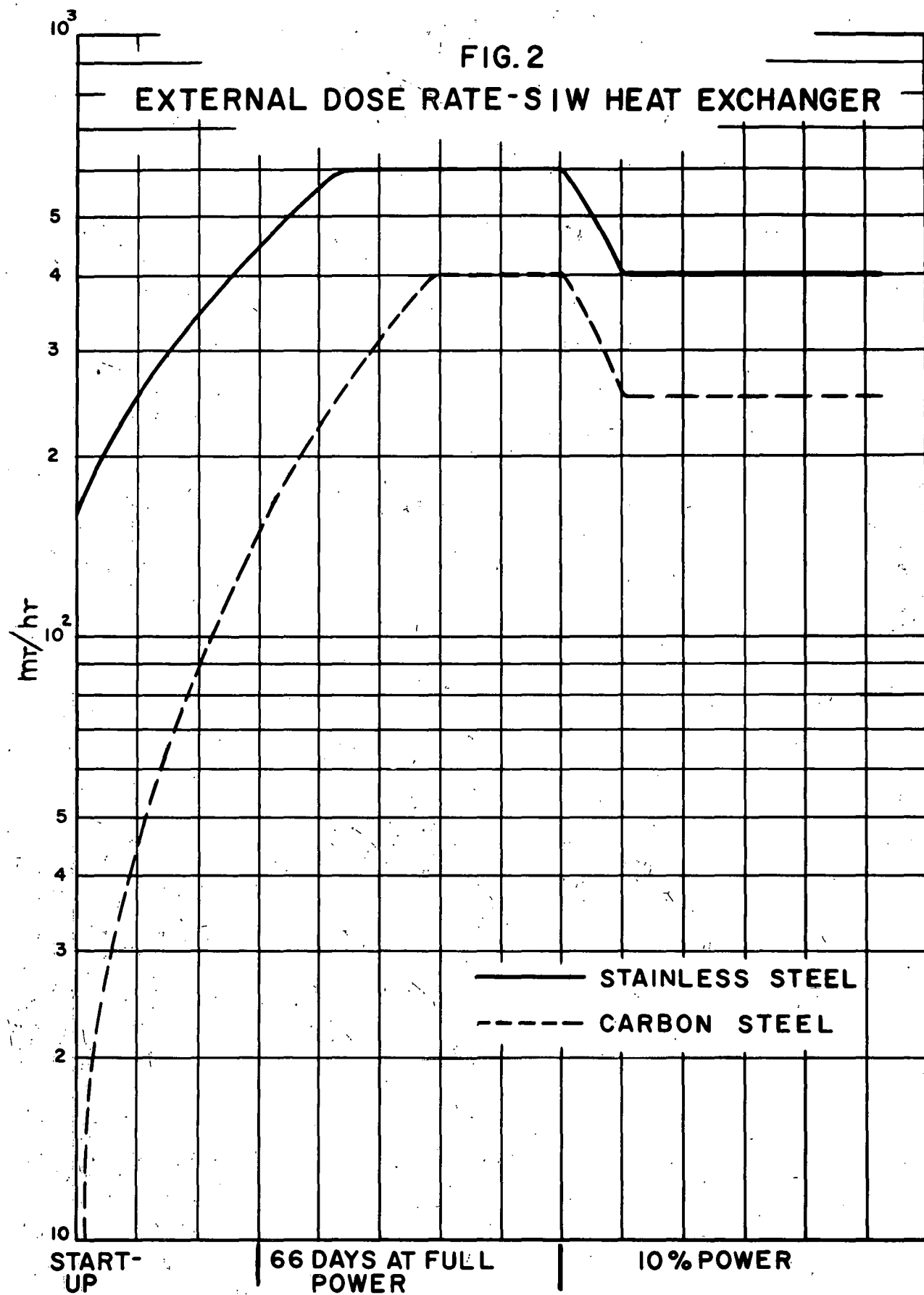
#### D. CARBON STEEL VERSUS STAINLESS STEEL

Significant differences at the S-1-W have been observed on both the deposition and the radiation emanating from deposits on the carbon steel boiler as compared to the stainless steel boiler. Figure 2 is a graphic presentation of activity observations<sup>(16)</sup> during operation for the two types of heat exchangers.

Deposition occurred in both heat exchangers and the external level was a function of power level. The carbon steel port heat exchanger had no activity at the beginning of the test so the build-up is from time zero. Of interest is the stainless steel heat exchanger, which already had prior deposition, yet the difference in level did not increase during subsequent runs.

Measurements on the two heat exchangers<sup>(16)</sup> for external dose rates and type of deposition are summarized in Table XVIII.





**TABLE XVIII. CARBON STEEL VERSUS STAINLESS STEEL  
HEAT EXCHANGERS**

Parameter	Carbon Steel	Stainless Steel
Nature of deposit	Loosely adherent	Tightly adherent
Thickness of deposit	500 mg/dm <sup>2</sup> on inlet to 70 mg/dm <sup>2</sup> on outlet	Thin, uniformly distributed
External dose rate	250 - 350 mr/hr	350 - 450 mr/hr
Internal dose rate	Inlet 2.5 - 10 r/hr Outlet 1 r/hr	Uniform - 1 r/hr

The carbon steel unit had a loosely bound non-uniform deposit and the deposit was larger in quantity than that in the stainless steel unit. Since the corrosion rate of carbon steel is higher than that of stainless steel, the larger amount of deposits is expected. The specific activity of the carbon steel deposits was lower than that for stainless steel, but the product of specific activity and quantity contributed to a larger total amount of overall activity than that for the stainless steel. Although the total carbon steel activity was larger than that of the stainless steel, its external dose rate was lower than that of the stainless steel heat exchanger.

Table XIX presents data on the corrosion rates of carbon and stainless steel with the relative amount of crud resulting from both of these heat exchangers. The higher corrosion rate for carbon steel overshadows the small total surface area sufficiently so that the chemical composition of the circulating crud may be controlled by the carbon steel corrosion product composition.

**TABLE XIX: EFFECT OF CARBON STEEL ON CIRCULATING CRUD**

<b>Material</b>	<b>Relative Surface Area</b>	<b>Corrosion Rate (mg/dm<sup>2</sup>/mo)</b>	<b>Amount of Crud(Relative)</b>
Carbon Steel	.23	30	6.9
Stainless Steel	.77	5	3.85

#### IV. POSSIBLE PHENOMENA IN ACTIVITY BUILD-UP

Various mechanisms have been proposed as possibly causing the activity build-up on primary system components. These include such phenomena as deposition, plating, and atom exchange. All of these, at this time of writing, can only be considered preliminary theories with insufficient evidence to support any particular one as the major phenomena. It is extremely doubtful that any conclusive mechanism will be formulated until more experimental results are obtained.

##### A. DEPOSITION

Early in 1956, data collected at S-I-W was interpreted<sup>(17)</sup> to imply that the specific activity of lightly adherent deposits on a removable sub-assembly was of the same order of magnitude as the specific activity of circulating crud. WAPD subsequently initiated a study on the activity transport phenomena in which additional quantitative data were obtained to further lend credence to these observations. In the quantitative study, the specific activity of the deposited material was studied and compared to values of circulating crud specific activity as shown in Table XX.<sup>(18)</sup>

TABLE XX SPECIFIC ACTIVITY OF CIRCULATING AND DEPOSITED CRUD

Isotope	Range, SA (dpm/mg) Circulating		Range, SA (dpm/mg) Deposited	
	Upper Limit	Lower Limit	Upper Limit	Lower Limit
Co <sup>60</sup>	$4.41 \times 10^6$	$5.11 \times 10^5$	$5.94 \times 10^5$	$1.56 \times 10^5$
Co <sup>58</sup>	$1.45 \times 10^6$	$1.63 \times 10^5$	$1.07 \times 10^5$	$3.83 \times 10^4$
Cr <sup>51</sup>	$15.4 \times 10^4$	$5.35 \times 10^4$	$1.64 \times 10^4$	$3.68 \times 10^3$
Fe <sup>59</sup>	$11.0 \times 10^4$	$3.99 \times 10^4$	$3.90 \times 10^4$	$7.12 \times 10^3$
Zr <sup>95</sup>	$13.5 \times 10^4$	$2.85 \times 10^4$	$2.12 \times 10^4$	$9.10 \times 10^3$
Hf <sup>181</sup>	$14.5 \times 10^6$	$1.57 \times 10^6$	$6.79 \times 10^5$	$1.76 \times 10^5$

Although these results appear to support this theory, the data was obtained by smear samples. The loosely bound material may be related to the circulation crud specific activity but whether or not the entire deposits follow this correlation, only extensive experimental work will determine.

In this same WAPD study it was stated an inverse relationship existed between activity crud level and crud specific activity.<sup>(19)</sup> The data is shown in Table XXI.

TABLE XXI RELATIONSHIP BETWEEN CRUD LEVEL AND CRUD SPECIFIC ACTIVITY

Crud Level (ppm)	Crud Specific Activity (cpm/mg)
0.13	$5.75 \times 10^6$
0.07	$1.08 \times 10^7$
0.028	$1.80 \times 10^7$
0.023	$3.75 \times 10^7$
0.02	$4.75 \times 10^7$
0.018	$2.75 \times 10^7$
0.018	$3.75 \times 10^7$
0.016	$4.75 \times 10^7$
0.016	$6.0 \times 10^7$
0.013	$6.75 \times 10^7$
0.010	$6.25 \times 10^7$

From the data, it appears that additional experimental data is needed to confirm this observation. Should specific activity be a controlling factor rather than total circulating activity which is the product of specific activity (dpm/mg) and crud level (mg/ml), and if this inverse relationship exists, the effectiveness of the purification system might be significantly reduced.

For the S-1-W system, the average increase in activity per  $\text{cm}^2$  during this study interval multiplied by the total surface area gives a value for total deposited activity which is about twice the amount of activity removed by the demineralizer in the same interval. Combined with the observation that

activity in the lower level reactor compartment increased by a factor of two in the same interval, this was interpreted to mean that the purification system and the system walls were about equally effective in removing activity during the run. This is the basis for the 0.5 factor used in the WAPD equation which will be discussed later.

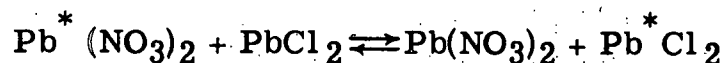
## B. EXCHANGE REACTIONS

### 1. Basic Phenomena:

The other phenomena which may be important to the activity build-up is exchange. Exchange is defined as a chemical reaction in which isotopes of an element interchange between one or more chemical forms of the element. The driving force for the reaction is entropy. Exchange reactions are classified into two major types, homogeneous and heterogeneous.

#### (a) Homogeneous Exchange:

An example of homogeneous exchange is the reaction between active lead nitrate and inactive lead chloride in water solution.

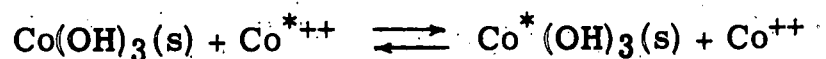


These reactions are often characterized by the reactants being evenly distributed in a single phase. All the atoms involved are chemically equivalent to one another and the reactions follows an exponential rate law. The actual exchange may be accomplished by several paths including dissociation, electron transfer, and atom transfer.

#### (b) Heterogeneous Exchange:

This type of exchange is exceedingly complex and takes place

where the reactants are in different phases and unevenly distributed. An example would be the exchange between solid cobalt hydroxide and radioactive cobalt in solution. (20)



Heterogeneous exchange reactions do not follow any simple rate law. The actual exchange may proceed in two steps. The initial step involves a rapid exchange at the phase interface. This is followed by a slow rate-determining step wherein the exchanged atoms are incorporated into a solid phase by the process of self-diffusion and/or recrystallization. The actual path will depend upon the particular system i. e., solid-solid, solid-liquid, solid-gas, etc.

The rate with which precipitated Ag Br exchanges with the component ions is different for the cation and anion. The interpretation for the slower  $\text{Br}^-$  exchange rate is that the rate controlling step is recrystallization. For  $\text{Ag}^+$  exchanging with the same precipitate, the increased rate is explained by more rapid incorporation into the solid phase as a result of recrystallization plus diffusion.

## 2. Evidence for Exchange:

The observation that the circulating crud and the deposited material have specific activities of the same order of magnitude may also be explained by an exchange mechanism. Preliminary information indicated that after



complete descaling, activity was found on the base metal itself, perhaps several atomic layers below the metal surface.

### 3. Additional Studies:

In reactor systems, the exchange may take place between a solid state dispersed in a liquid and solid metal surfaces. No published studies were found on this type of exchange. However, the exchange of  $\text{Co}^{60}$  in solution with various metal surfaces has been investigated. (21) In this study, small metal test specimens were placed in a water solution of  $\text{Co}^{60} \text{SO}_4$  (50,000 cpm per cc). The reaction vessel was so designed that oxygen could be excluded. After various intervals of time, test specimens were removed from the active solution, washed, dried, and counted. Some data from this study are presented in Table XXII.

**TABLE XXII HETEROGENEOUS EXCHANGE OF  $\text{Co}^{60}\text{SO}_4$  IN SOLUTION  
WITH METAL SURFACES**

Specimen	Surface Treatment	Time of Exchange	Activity (cpm)		Atom Layers/cm <sup>2</sup>	
			O <sub>2</sub> Present	O <sub>2</sub> Absent	O <sub>2</sub> Present	O <sub>2</sub> Absent
Co	Etched	1 min.	2,600	--	.26	--
		5	20,300	9,600	2.03	0.92
		30	32,000	11,800	3.20	1.18
		120	50,100	16,000	5.01	1.60
		1,440	52,200	17,700	5.22	1.80
		2,880	40,500	20,300	4.05	2.03
Co	Ground 000 (thermally etched)	120	53,300	24,200	5.33	2.42
Co	Cold rolled (etched)	120	44,500	10,500	4.45	1.05
Ni	Cold rolled (etched)	120	47,000	2,350	4.70	.235
Ni	Cold rolled (etched) HF	120	49,000	8,600	4.90	.86
Fe	Cold rolled	15	70,000	32,200	7.0	3.22
Fe	Cold rolled	120	103,400	53,400	10.3	5.34
Fe	Ground 000	120	106,000	12,800	10.6	1.28
18-8 SS	Etched	120	550	1,700	.053	.17
	Etched air 5 min.	120	340	--	.034	--
	Etched air 1 day	120	190	--	.019	--
	Etched	120	920	--	.09	--
	Ground	120	16,500	140	1.64	.014

The following conclusions were drawn from this data:

1. The position of the metal ion in solution in the E. M. F. Table relative to the solid phase metal markedly affects the amount of exchange. Metals more noble than the metal ion in solution acquire less activity. Iron, which has a potential of  $\sim 0.44$  volts, is the only metal which is less noble than cobalt (potential  $\sim 0.28$  volts) and it acquired the greatest amount of activity.
2. The influence of  $O_2$  depends upon the nature of the oxide film formed. If the oxide film is not protective, oxygen increases the amount of activity in the metal. A protective oxide film markedly decreases the activity by limiting the areas where electrolysis can take place.
3. Radioactivity acquired by metal surfaces is governed by local anodes and cathodes of the metal surface. This is shown by the marked increase in the activity of the ground stainless steel sample.

Of importance is the fact that by this electrolytic mechanism it is not necessary to have isotopes of the same element. However, if these experiments could be extrapolated to reactor conditions, it would be expected that cobalt activity on the carbon steel exchanger would have been much higher than on the stainless steel. Since this is not observed, this can not be the only mechanism operative. On the other hand, the presence of  $Co^{60}$  activity on the carbon steel boiler indicates that this mechanism may be a factor to consider.

## V. PARAMETERS AFFECTING DEPOSITION

As a result of the literature survey and personal contacts, many different parameters are believed to contribute to the activity build-up. As would be expected in attempting to define a new phenomena, considerable differences of opinion exist on what parameters play a significant part in contributing to the activity.

### A. PURIFICATION

All pressurized water reactors, either in operation or being designed, employ a by-pass purification system to maintain high purity water. Although equipment variations exist in the different by-pass purification systems, they are basically very similar. Some systems employ a high pressure by-pass system while others, as in the APPR, employ a low pressure by-pass purification. It is doubtful whether the differences in pressure will affect the activity build-up in any respect.

The basic premise for the purification system from the standpoint of the activity build-up is that a reduction in the total activity will reduce the total amount of build-up. Consequently, the more effective the purification, the higher reduction in the build-up of activity. Reducing the total circulating activity would appear to reduce the total build-up on the primary system components. Many investigators however, are not in agreement with this last statement. They contend that the specific activity (dpm/mg) is the prime important factor and the total activity is of no major consequence. It has been stated even by some that the purification system is of no value in relation to the activity build-up program. Obviously, considerable confusion exists on the effect of this one parameter alone.

In the design of a purification system, unfortunately the location of the blow-down line may be given only cursory consideration. If the blow-down line is inserted in an area where representative portions of the main primary water are not removed, the purification effectiveness can be reduced. A determined effort should be made to place the blow-down line so as to remove representative samples from the primary loop.

Since many of the corrosion products have been found to be insoluble, the prime factor in the purification system is the filtration ability. Its ability to remove the insoluble materials may be related directly to its ability to remove activity. Studies were performed to show the effectiveness of filtration only in removing activity. Table XXIII presents some of this data. (22)

TABLE XXIII EFFICIENCY OF FILTER FOR REMOVING ACTIVITY

	Upstream	Downstream	Filter Efficiency (% Removed)
Long t 1/2 (cpm/ml)(120h)	$1.5 \times 10^2$	$0.6 \times 10^2$	60
Short t 1/2 (cpm/ml)(5 min)	$3.3 \times 10^4$	$2.8 \times 10^4$	15
Specific Activity Crud (cpm/mg)	$8-10 \times 10^5$	$2 \times 10^5$	77
Concentration Crud (ppm)	0.12	0.05	58

Of interest in this study is that the major portion of the short half-lived nuclides were not associated with the insoluble material whereas the long-lived activity was at least 60% associated with insoluble products. A filter could conceivably serve as the entire purification system if the porosity were fine enough to remove 100% of the insoluble material with acceptable pressure drops.

It is interesting to speculate on the observed change in specific activity noted.

Either the finer particles, which are not removed by the filter have less activity per unit weight or the filter itself is adding nonactive insoluble products to the system.

The most commonly used method in reactor operations for removing impurities from water is demineralization. Although a demineralizer is designed to remove soluble activities by exchange, it also serves as a very efficient filter. The efficiency of the demineralizer in removing activity is determined by the decontamination factor which is the ratio of influent to effluent activity.

Data is presented in Table XXIV on the performance of the demineralizer at the APPR-1 during the 700 hour test. <sup>(23)</sup> At the APPR-1, the demineralizer acts both as a filter and an exchanger and consequently activity is removed whether it is associated with soluble or insoluble products.

TABLE XXIV DECONTAMINATION FACTORS DURING APPR-1  
700 HOUR TEST

Date	Equivalent Full Power Hours	Upstream cpm per cc	Downstream cpm per cc	Decontamination Factor
June 4	154.5	18,350	206	89
June 10	220.8	43,100	520	83
June 11	240.9	46,700	229	204
June 11	244.6	55,900	337	166
June 14	319.3	49,250	436	113
June 22	490.9	83,700	258	325
June 26	588.6	56,700	160	354

## B. HYDROGEN EFFECT

Control of the corrosion release products will control the rate at which activity enters the system. At the S-1-W, the  $H_2$  concentration<sup>(24)</sup> was found to affect the activity level as shown in Table XXV. A low  $H_2$  concentration apparently increases the short half-lived circulating activity but decreases the specific activity of the crud. Other operating data indicates that hydrogen affects the nature of soluble and insoluble corrosion products, particularly with respect to crud filterability.

TABLE XXV EFFECT OF HYDROGEN ON SYSTEM ACTIVITY  
(55-60% Power)

Activity	Hydrogen Concentration	
	100 cc/kg	10 cc/kg
Short-lived water activity, c/min/ml	$3.5 \times 10^4$	$6-10 \times 10^4$
Spec. Activity of crud, c/min/mg	$8-10 \times 10^5$	$4 \times 10^5$
Long-lived activity in water	Varies as the crud level	

## C. EFFECT OF pH

In conventional boilers it is standard practice to maintain high pH levels to minimize both corrosion and deposition of magnetite on system surfaces such as boiler tubes. Corrosion in condensate systems has been minimized by raising the condensate pH. Recent investigations at KAPL and Batelle<sup>(25)</sup> have shown that ferrous hydroxide decomposition is not the rate controlling step, suggesting that the reaction of metal with oxygen or water is controlled by a diffusion process.

The role of pH on carbon steel corrosion has been demonstrated by recent tests<sup>(26, 27)</sup>. Figure 3 compares the descaled corrosion rate of carbon steel in high temperature water at neutral and high pH. High pH reduced the corrosion rate of all carbon steels tested to about 40 mg/dm<sup>2</sup>/mo after 1000 hours exposure. This is equivalent to approximately 0.0002 inches of steel metal loss per year.

Early observations in test set-ups such as those at Chalk River, indicated that pH control produced favorable results. Considerable effort has been expended by the naval nuclear propulsion program to determine the effect of pH on corrosion in a pressurized water reactor primary system. Generally, high pH (10 to 11) has been found to have a beneficial effect in reducing the quantity of corrosion products released in an in-pile stainless steel system. This of course would affect the total activity deposited on primary system components.

The yield of transportable corrosion products<sup>(28)</sup> is much lower at high pH than with neutral water and high pH inhibits the deposition of crud. This inhibiting effect is shown in Figure 4. Compared to neutral water, the corrosion film is quite thick.

Operation of the S-1-W has shown that high pH not only reduces the quantity of corrosion products released to the system, but those that are released are more filterable. These in-plant tests also demonstrated that water dissociation was not increased and that mechanism performance was not affected. Except for an accelerated corrosion on chrome-plate when oxygen levels were above 0.1 cc/kg, operation at high pH has been quite beneficial on general system performance.



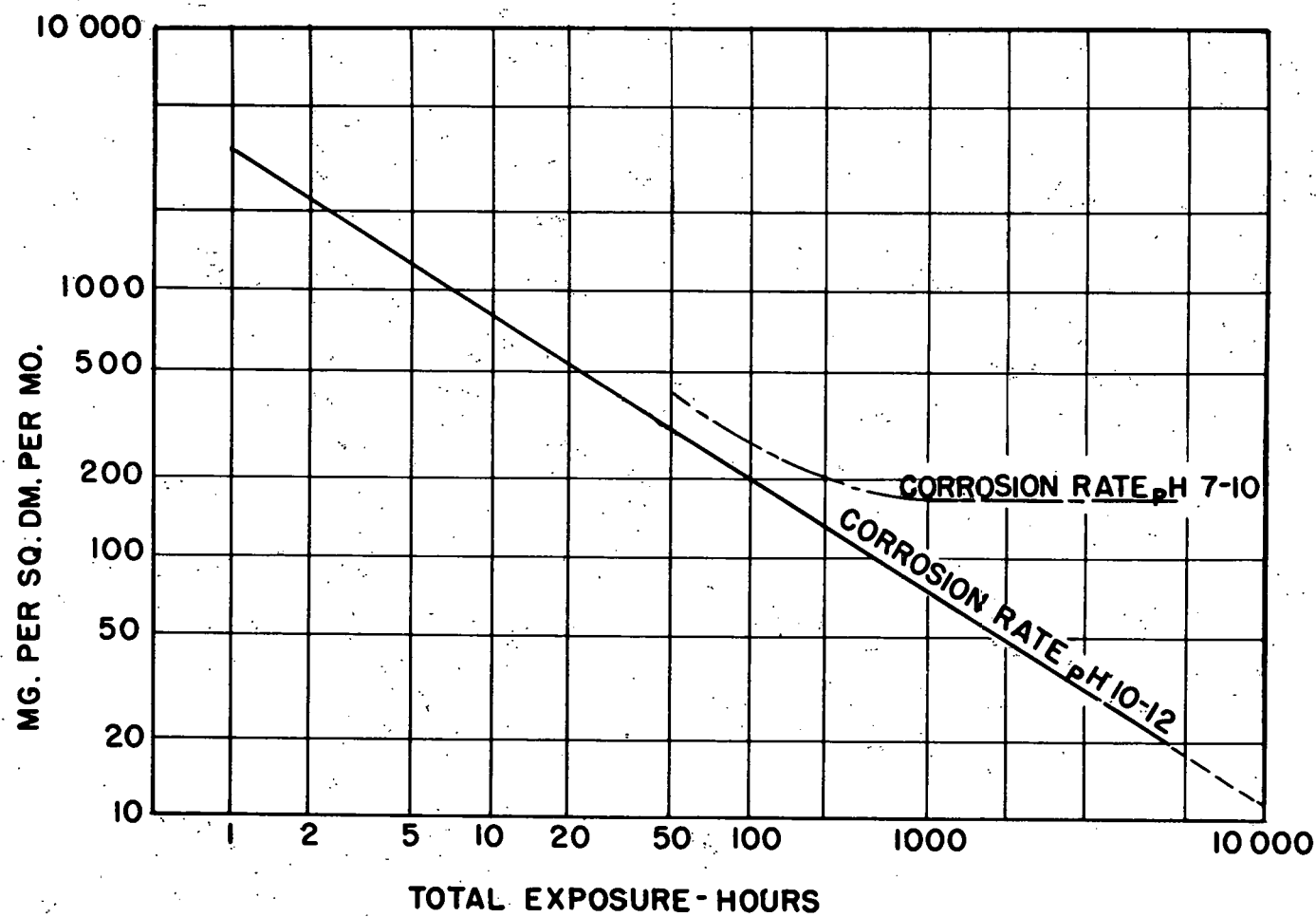
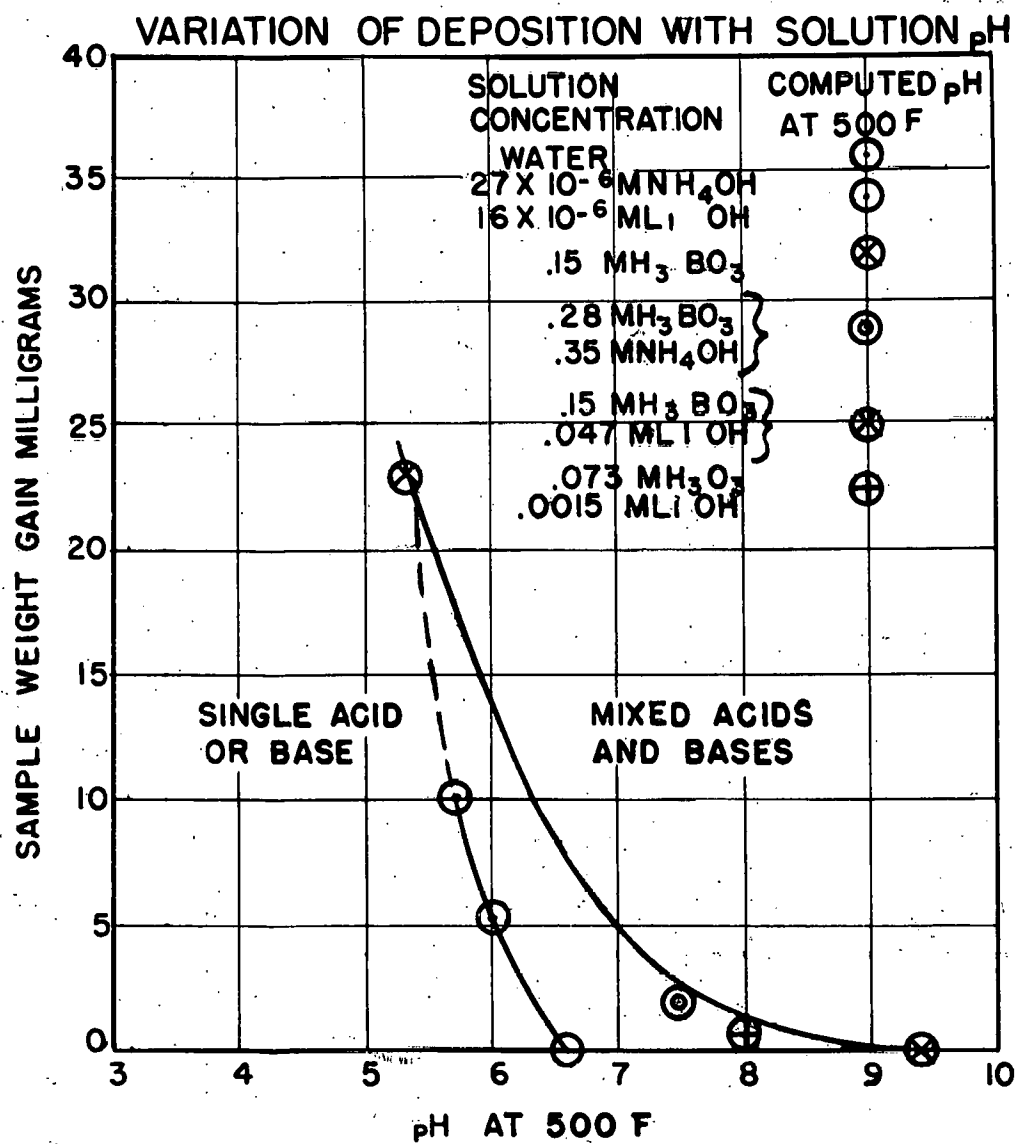


FIG. 3 INSTANTANEOUS DESCALED CORROSION RATE OF CARBON STEEL IN HIGH TEMP. WATER.

FIG. 4



#### D. EFFECT OF IN-PILE VERSUS OUT-OF-PILE AREA

The relative importance of "in flux" material compared to material originating in "out of flux" regions towards the total system activity has not been established. WAPD and KAPL disagree on their relative importances. WAPD reports that "The contribution to deposited activity from activation of circulating crud produced by corrosion of non-irradiated surfaces has been neglected." On the other hand, KAPL, in connection with the SAR project, is considering "out of flux" regions as a significant, if not controlling, factor.

The importance of the area ratio is emphasized in the selection of materials of construction. Since cobalt is a chief offender in the activity build-up, its elimination from all of the materials of construction would essentially eliminate the  $\text{Co}^{60}$  presence. This approach will be extremely costly.

If it appears that the WAPD statements regarding the contribution of "out of flux" areas is valid, the elimination of cobalt impurities in the high flux areas would essentially eliminate the  $\text{Co}^{60}$  problem. Should it be the KAPL assumption that is valid, then the ratio of the "in flux" to "out of flux" area is important. Parameters necessary to evaluate the contribution of each area include release rates, resident times in high flux areas, and redeposition rates. The ratio of the specific activity of the released material from the "out of flux" area to the specific activity of the released material from the core area may be the controlling factor. Little data is available, if any, to predict what specific activities would result from the different areas.

#### E. DEPOSIT THICKNESS

The crud deposits that have been reported describe this material

as a black adherent film of magnetite structure. It is of uniform thickness and in-flux surfaces have been found to have a thicker layer than that deposited on out-of-flux surfaces for Zircaloy. In-pile specimens were reported to have a coating four times thicker than the out-of-flux surfaces.<sup>(29)</sup> Although the work was reported on zirconium coupons, it appears that a similar effect occurred on stainless steel. A thicker deposit occurred on zirconium than on stainless steel. Water conditions such as gas concentrations, pH, and temperature appear to markedly effect the deposits.

Based on in-pile loop studies, KAPL<sup>(30)</sup> believes that an active equilibrium thickness does occur and that after a certain time period this active equilibrium thickness of .25 mg/cm<sup>2</sup> is obtained. Although the thickness reaches an equilibrium its specific activity continues to build up with time.

WAPD does not agree that an equilibrium active thickness is reached and that deposits do continue to build up with time. In their calculations<sup>(31)</sup> for predicting the activity, a thickness of .42 mg/cm<sup>2</sup> was assumed after five years of PWR operation.

#### F. SPECIFIC ACTIVITY

The specific activity of the circulating deposits is important whether the phenomena be deposition or atom exchange. If the material being deposited or replaced on a surface rises from crud with a higher specific activity, the inevitable outcome is a higher build-up. The total amount of activity, is the product of specific activity (dpm/mg) and total amount of material (mg). If the specific activity of circulating and deposited crud are of the same order of magnitude and if the rate of build-up of deposit thickness is known, a simplified

prediction of the total extent of activity build-up would result. From the data presented between the levels of the circulating crud and its specific activity, it is too premature to definitely state the relationships between the two.

#### G. EFFECT OF FLOW RATE

The effect of coolant flow rate on deposition is important. Unfortunately, data available to indicate quantitatively what its effect will be is lacking. In stagnant areas, the deposition of crud would be expected to be greater than in the high flow areas. As an example of the deposition which might occur at stagnant areas, reference is made to Table XV of dose rates after the 700 hour test at APPR-1. In the pressurizer dead leg area, essentially no flow exists, and the large deposits resulted. These so-called crud traps probably exist in many areas of all reactor designs.

#### H. OTHER PARAMETERS

In surveying the literature and as a result of personal contacts, very little information was available on the following parameters:

1. Effect of temperature.
2. Effect of surface finish.
3. Effect of pressure.

## VI. THEORETICAL PREDICTIONS

### A. GENERAL

All present methods of theoretically predicting activity levels depend upon the simple material balance i. e.,

Rate of accumulation = rate of formation - rate of decay.

A general qualitative discussion of the phenomena involved will be given and two representative equations derived and discussed.

#### 1. Rate of Formation

The data which forms the basis upon which various phenomena have been postulated were discussed in a previous section. In this section, the parameters are separated to determine their individual effect on the activity problem.

As previously mentioned, the two more important mechanisms by which radioactive nuclides are added to the system are release of active nuclides from in-flux surfaces and activation of corrosion products released from structural materials while residing in and circulating through flux regions. For products released from in-flux surfaces, the specific activity (dpm/mg) approaches its saturation value according to the following relationship:

$$A = N_0 \sigma \phi (1 - e^{-\lambda t}), \text{ where:}$$

A =	number of active atoms
N <sub>0</sub> =	number of parent atoms
σ =	activation cross section
φ =	neutron flux
λ =	decay constant
t =	time exposed to flux

For a given reactor, the  $(\phi)$  and number of parent nuclides are fixed (assuming constant power level and neglecting burnup of the parent nuclides). Consequently, the effort to reduce activity from the in-flux area is primarily a function of control of the release rate for a particular material of construction. By careful selection of the materials of construction it is possible to limit the number of parent atoms. The other two variables are nuclear properties of the parent nuclide and hence cannot be altered.

For short half-lived nuclides, the value of  $(1 - e^{-\lambda t})$  essentially become unity and short half-lived nuclides reach their saturation activity rapidly. Conversely, the short-lived nuclides decay more rapidly and do not offer the long-range accumulation problem associated with long-lived nuclides.

For parent nuclides released from out-of-flux structural materials, it is felt that deposition on in-flux surfaces, especially fuel elements, will have a pronounced effect on activity levels. With time in the high flux area, the nuclides will approach their saturation activity and consequently when released will make a larger contribution to the activity problem.

## 2. Rate of Removal

The other factor in the material balance that determines system activity levels is the rate of removal of activated products. The active material can be removed from the system by four recognized mechanisms: purification, deposition, atom exchange, and decay. Of these four mechanisms, only decay and rate of purification have been considered to-date in most theoretical approaches. As discussed previously, an empirical deposition factor of 0.5, however, is used by WAPD to make a correction for deposition. All of the present equations assume

a known fraction of the activities are removed by the purification system. Removal of the radioactivity due to decay is the only mechanism that can be clearly defined.

Released nuclides, are principally insoluble and, for the most part, activity associated with water is mainly due to insoluble transport products. Before the purification system can remove all the insoluble material, deposition often can occur. Thus the actual effectiveness of the purification system with respect to controlling radioactivity build-up has not been clearly established. Assuming 100% removal as is usually practiced is certainly not true as indicated in certain equations. The contribution of purification towards controlling activity build-up will be a definite function of the magnitude of the deposition and exchange phenomena.

The remaining two mechanisms for removing activity from the system are deposition and exchange. Until only recently, both factors were ignored in predicting system activity levels and even now arbitrary corrections are applied to existing equations in an attempt to more nearly approach experimental values. No published equations attempt to include exchange effects.

#### B. WAPD EQUATION

The derivation of the equation which currently is used at WAPD<sup>(31)</sup> is given below. The differential form for accumulation in a core is:



$$\frac{dN_b^c}{dt} = Q_a^c \sigma_a \phi_e - \lambda_b N_b^c \quad \dots (1)$$

where:  $Q_a^c$  = concentration of atoms being activated by reaction

$a(n, \gamma)b$ , (atoms/gm alloy)

$\sigma_a$  = thermal neutron cross section ( $\text{cm}^2/\text{atom}$ )

$\phi_e$  = effective thermal neutron flux ( $\text{n}/\text{cm}^2\text{-sec}$ )

$N_b^c$  = concentration of atoms of isotope b formed in core by  
by a ( $n, \gamma$ )b (atoms/gm alloy)

$\lambda_b$  = decay constant of b ( $\text{sec}^{-1}$ )

t = time (sec)

The solution for the first order linear differential equation is:

$$N_b^c = \frac{Q_a^c \sigma_a \phi_e}{\lambda_b} (1 - e^{-\lambda_b t}) \quad \dots (2)$$

Since the chief interest is in the rate at which activity accumulates on primary components, the analysis continues as follows:

$$\frac{dN_b^c}{dt} = \frac{0.5 C_c A N_b^c}{A_s} - \lambda_b N_b^s \quad \dots (3)$$

where: C = overall corrosion rate ( $\text{gm}/\text{cm}^2\text{-sec}$ )

$A_c$  = area exposed to water in core ( $\text{cm}^2$ )

$A_s$  = Area of primary system - deposition area ( $\text{cm}^2$ )

$N_b^s$  = atoms of b deposited in system ( $\text{atoms}/\text{cm}^2$ )

0.5 = assumed ratio of deposited crud to total system crud

If the half-life of b is small, ( $1 - e^{-\lambda_b t}$ ) approaches 1 and  $N_b^c$  is a constant. Substituting the value for  $N_b^c$  from equations 2 and solving the

resulting first order linear differential equation gives:

$$\lambda_b N_b^c = \frac{0.5 C A_c Q a^c \tau_a \phi e}{\lambda_b A_s} (1 - e^{-\lambda_b t}) \quad (4)$$

Once again  $(1 - e^{-\lambda_b t}) \cong 1$  since the half-life is small.

$$\lambda_b N_b^s = \frac{0.5 C A_c Q a^c \tau_a \phi e}{\lambda_b A_s} \quad (5)$$

However, if the half-life is not small, equation 2 must be substituted as a function of time into equation 3. The resulting differential equation is still a linear first order but the solution becomes:

$$\lambda_b N_b^s = \frac{0.5 C A_c Q a^c \tau_a \phi e}{\lambda_b A_s} (1 - \lambda_b t e^{-\lambda t} - e^{-\lambda_b t}) \quad (6)$$

These equations of WAPD are the only ones which presently make an effort to predict deposited activity.

Some interesting observations of the above equation are listed below:

1. The formation of nuclides in the core and the deposition of nuclides throughout the system is expressed on a weight basis; thus, essentially specific activity (dpm/mg) is considered.
2. The overall corrosion rate is considered equal to the release rate. Since the corrosion rate is higher than the release rate, a source of error exists, particularly for long irradiation time.
3. Deposition is considered uniform in the flux as well as out of

the flux region. Data has been presented indicating that there is a thicker layer on "in-flux" regions than on the rest of the system.

4. Nuclides result only from irradiation of structural materials located in "in-flux" regions. It is very probable that target nuclides which are released from "out-of-flux" regions and subsequently deposit in "in-flux" areas are also a major contributor to system activities.

5. Exchange reactions are not considered.

6. The 0.5 ratio of deposited crud to total crud is an arbitrary value.

7. If  $t = 0$  is assumed as any time other than the actual start-up time, account would have to be taken of the active nuclides which remain in the system as well as those actually formed in the new irradiation interval.

8. Only  $(n - \gamma)$  reactions are considered. An indirect specific method was given which makes it possible to include the observed  $(n-p)$  product  $Co^{58}$ .

Even with these apparent shortcomings, this equation is probably the best presently available. Observed deposited activities can still at best be only approximated within perhaps an order of magnitude.

### C. KAPL EQUATION

The most recent equation used by KAPL<sup>(30)</sup> is as follows:

$$\lambda N_c = \sigma \phi_c N (1 - e^{-\lambda t})$$

where:

$N_c$  = number of active nuclei

$N$  = number of target nuclei

$\sigma$  = thermal absorption cross section

$\lambda$  = decay constant of active nuclei

$t$  = time of operation

$\phi_c$  = thermal flux in the core

This equation is, of course, identical to the one derived in the previous section. The next step is to establish an equivalent area exposed to an equivalent flux.

This is shown below:

$$\sum_{s=1}^{s=N} \phi_s A_s = \phi_c A_{\text{core}}$$

where

$\phi_s$  = average thermal flux over stainless steel area  $A_s$ .

$\phi_c$  = core thermal flux

$A_{\text{core}}$  = pseudo area of stainless steel which can be considered as in the core flux

The fraction of the total system corrosion of target nuclei which is already active is (located in "in-flux" regions):

$$f = \sigma \phi_c \frac{A_{\text{core}}}{A_{\text{total}}} \frac{1}{\lambda} (1 - e^{-\lambda t}) \quad \dots (8)$$

where:

$A$  = total stainless steel area exposed to primary coolant

The differential equation is then:

$$\frac{dN_w}{dt} = \frac{\sum \phi_i t_i}{T} Neq \sigma - \frac{P N_w}{T} + \frac{P}{T} f Neq - \lambda N_w \quad \dots (9)$$

where:  $\sum \phi_i t_i$  = sum of products of average flux in each region and time in region in each primary coolant cycle

$T$  = total cycle time (sec)

$P$  = fraction of impurities removed by demineralizer per cycle

$Neq$  = concentration of mother nuclei in coolant

The solution of (9), (a first order linear differential equation) is:

$$N_w = - \left[ \frac{\sum \phi_i t_i + \frac{P}{\lambda} \phi_c \frac{A_{core}}{A_{total}} - \frac{\phi_c}{\lambda} \frac{A_{core}}{A_{total}} e^{-\lambda t}}{P + \lambda T} \right] Neq \sigma e^{-\left(\frac{P}{T} + \lambda\right)t} + \left[ \frac{\sum \phi_i t_i + \phi_c \frac{P}{\lambda} \frac{A_{core}}{A_{total}}}{P + \lambda t} \right] Neq \sigma - \frac{\phi_c}{\lambda} \frac{A_{core}}{A_{total}} \sigma Neq e^{-\lambda t} \quad \dots (10)$$

This equation can be simplified by assuming that:  $N_w = 0$  ,  $t = 0$

and that  $\left(\frac{P}{T} + \lambda\right)t \gg 1$

$$N_w = \frac{\left(\sum \phi_i t_i + \phi_c \frac{P}{\lambda} \frac{A_{core}}{A_{total}}\right) Neq \sigma}{P + \lambda t} - \frac{\phi_c}{\lambda} \frac{A_{core}}{A_{total}} \sigma Neq e^{-\lambda t} \quad \dots (11)$$

The assumption leading to the simplification was shown to be valid for all times greater than twenty hours.

An examination of the equation and derivation leads to the following observations. Deposition is ignored entirely with the following consequences:

(1) The demineralizer is assumed to see all the released corrosion products and remove a known fraction periodically. Data has been presented to show that this is not a valid assumption.

(2) The target nuclei released from "out-of-flux" regions do not deposit on "in-flux" surfaces but become irradiated only as a result of passing through the core while suspended in primary coolant.

(3) If this ideal condition were the case, no accessibility problem would exist since all activity could be removed by draining the primary water.

Other observations are as follows:

(1) The equation is solved on a volume basis (dpm/ml) and, consequently, increasing purification flow rates reduce the circulating activity.

(2) This equation gives some weight to target nuclei originating elsewhere in the system and activated during circulation.

(3) As in the WAPD equation, the corrosion rate is assumed equal to the release rate.

(4) The equation is strictly valid only if (t) is the actual time of start-up. No provision is made for activity remaining after a decay period which would have to be added to the activity found during the new time interval.

(5) No (n, p) reactions are considered taken.

(6) Exchange reactions are not considered.

From the discussion of these two equations, many shortcomings in predicting system activity levels exist. Only the WAPD equation makes any effort to take into account activity deposition which is probably the major cause of the accessibility problem. These shortcomings can be eliminated only by an

experimental program in conjunction with a continuous theoretical effort.

#### D. PWR PREDICTED ACTIVITY LEVELS

Predictions with the WAPD equation have been made regarding the possible activity build-up on the PWR. <sup>(31)</sup> The results of this work is shown in Tables XXVII, XXVIII and Figures 5 and 6. In their calculations, overall corrosion rates for all materials except Type 410 stainless steel was 2 mg/dm<sup>2</sup>/mo. As can be seen, the estimate for cobalt activity is the dominating activity.

TABLE XXVI. CONTRIBUTION OF INDIVIDUAL PWR CORE MATERIALS TO DEPOSITED ACTIVITY

Core Material	% of Indicated Activity Originating in Material			
	At 1 Month		At 5 Years	
	Total	Co <sup>60</sup>	Total	Co <sup>60</sup>
304 Stainless Steel	13.2	13.8	13.7	13.8
Inconel - X	1.5	5.0	5.0	5.0
Stellites	17.3	76.4	74.5	76.4
Haynes	0.1	4.7	4.6	4.7
Hafnium	67.8	-	2.2	-

Other core materials (17-4 PH stainless steel, Type 410 stainless steel, chromium, and gold) have a negligible contribution to the deposited activity.

TABLE XXVII. CONTRIBUTION OF INDIVIDUAL RADIONUCLIDES TO CALCULATED TOTAL DEPOSITED SPECIFIC ACTIVITY

Nuclide	Percentage of Total Activity Due to Indicated Nuclide				
	1 Month	2 Months	6 Months	1 Year	5 Years
Cr <sup>51</sup>	2.9	1.9	1.0	0.5	0.1
Mn <sup>56</sup>	5.2	1.7	0.4	0.1	0.1
Fe <sup>59</sup>	2.8	2.7	1.9	1.0	0.1
Co <sup>58</sup>	2.0	2.2	2.0	1.4	0.1
Co <sup>60</sup>	19.0	24.5	45.5	70.4	97.6
Hf <sup>175</sup>	9.5	10.1	9.5	6.2	0.5
Hf <sup>181</sup>	58.5	56.9	39.7	20.4	1.6
	99.9 *	100.0	100.0	100.0	100.0

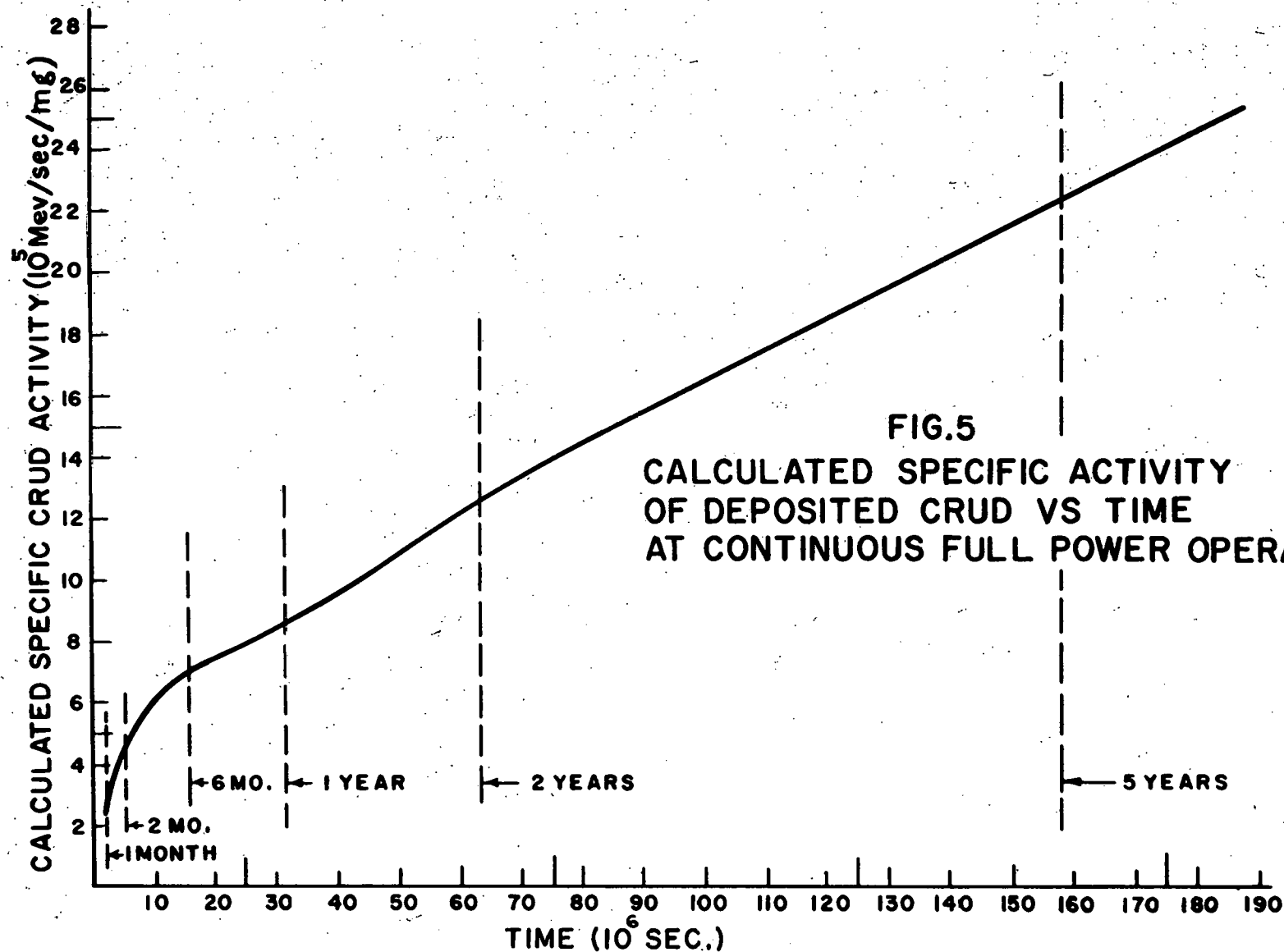
\* The remaining 0.1% in this case is due to minor amounts of Cu<sup>64</sup>, Ni<sup>65</sup>, Ta<sup>182</sup>, W<sup>185</sup>, and W<sup>187</sup>.

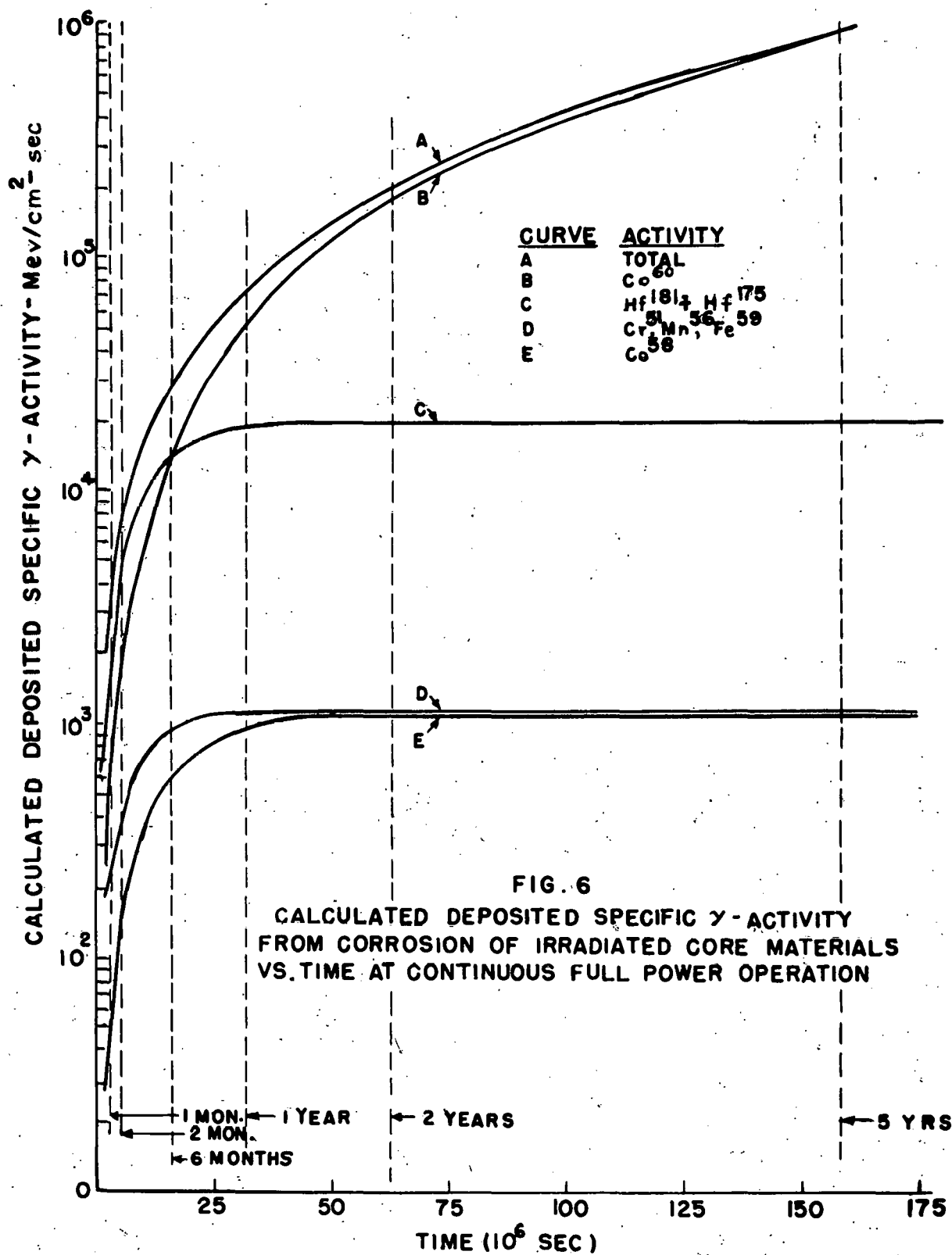
The table is based on maximum calculated hafnium activities.

TABLE XXVIII. CALCULATED CRUD BUILD-UP AND ACTIVITY DUE TO CRUD IN PWR AS A FUNCTION OF TIME AT CONTINUOUS FULL POWER OPERATION

Time	Total Amount of Deposited Crud (mg)	Total Deposited Crud Activity (Mev/sec)	Specific Activity of Deposited Crud (Mev/sec/mg)
1 month	$3.88 \times 10^5$	$1.12 \times 10^{11}$	$2.89 \times 10^5$
2 months	$7.76 \times 10^6$	$3.45 \times 10^{11}$	$4.45 \times 10^5$
6 months	$2.33 \times 10^6$	$1.63 \times 10^{12}$	$7.00 \times 10^5$
1 year	$4.66 \times 10^6$	$3.99 \times 10^{12}$	$8.56 \times 10^5$
2 years	$9.31 \times 10^6$	$1.16 \times 10^{13}$	$1.25 \times 10^6$
5 years	$2.33 \times 10^7$	$5.16 \times 10^{13}$	$2.21 \times 10^6$







KAPL <sup>(32)</sup> estimated the activity build-up for the S-3-G which utilizes zircaloy clad elements. The results are shown in Tables XXIX , XXX and Figure 7. Again the importance of cobalt is emphasized.

TABLE XXIX. WATER ACTIVITY IN DIS/SEC/CC AFTER E. F. P. H.

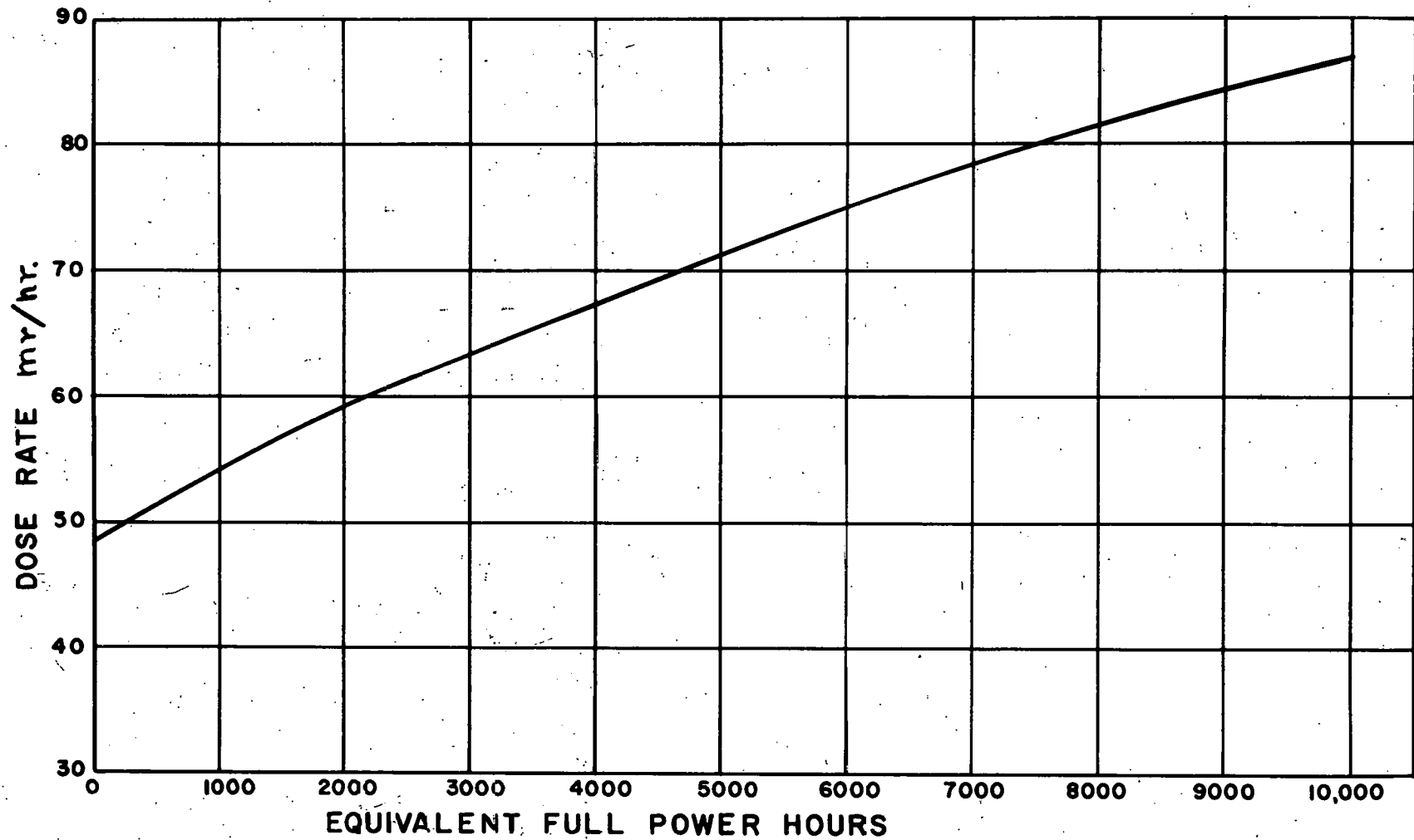
Element	100	500	1,000	3,000	6,000	10,000
Fe <sup>59</sup>	.56	1.6	2.5	4.4	4.9	5.0
Co <sup>58</sup>	.16	.7	1.3	2.8	3.6	4.0
Zr <sup>95</sup>	.04	.18	.31	.64	---	---
Hf <sup>181</sup>	.37	1.7	2.8	5.1	---	---
Co <sup>60</sup> (original)	.3	1.3	2.6	7.6	14.4	23.4
Co <sup>60</sup> (modified)	9.0	39.0	69.0	132	159	165

TABLE XXX. DOSE RATES, 15 MINUTES AFTER SHUTDOWN AFTER 2700 E. F. P. H.

Element	Water Activity dis/sec/cc	Dose rate mr/hr	Surface Activity dis/sec/cm	Dose Rate mr/hr	Total Dose rate, mr/hr
Fe <sup>59</sup>	4.2	.017	$2.1 \times 10^3$	2.9	3.0
Co <sup>58</sup>	2.6	.007	$1.3 \times 10^3$	1.2	1.2
Zr <sup>95</sup>	0.6	.011	$3.1 \times 10^2$	.278	.29
Hf <sup>181</sup>	5.0	.0006	$2.3 \times 10^3$	.805	.81
Co <sup>60</sup>	6.9	.06	$3.5 \times 10^3$	10.3	10.4

FIG.7

S3G/S4G PRIMARY COOLANT PIPING DOSAGE 15 MINUTES  
AFTER REACTOR SHUTDOWN



## VII DECONTAMINATION

Excessive radiation dose rates from components of a pressurized water system pose serious operational and maintenance hazards. Such a condition could be brought about by one of two means or very possibly a combination of both.

These are:

1. Deposition of activated nuclides (corrosion products, coolant impurities, etc.) on component surfaces during normal operation.

2. A sudden release of fission products from a ruptured fuel element.

As yet, no method has been found to control or eliminate either of the above mentioned phenomena. This being the case, the alternate approach of removing activity from system components has been the subject of much investigation. These investigations have centered around the hope of finding a solution that when circulated through a loop, would remove activated deposits from component surfaces without producing any deleterious effects on the individual loop components.

### A. INVESTIGATIONS TO-DATE

The majority of information presently available pertaining to decontamination resulted from studies conducted by WAPD. Their work has been divided into two general categories:

- I Laboratory and bench-scale investigations.
- II Full-scale trials on equipment and plants.

A description of progress to date on each phase of this work follows:

## B. LABORATORY AND BENCH-SCALE INVESTIGATIONS

Solution (A) (See Table XXXI) has been tested for  $\text{UO}_2$  and fission product removal<sup>(33)</sup> in a small scale loop (Bettis Loop No. 4, Runs 1, 2, 3, 5, 6). Loop hot filter decontamination factors of 10 were obtained in 20 to 25 hours when the loop was used in a feed-and-bleed once-through procedure (Runs 1, 2). Feed-and-bleed recirculation runs (3, 5, 6) yielded decontamination factors of about 5 within 30 to 35 hours.

TABLE XXXI. DECONTAMINATION SOLUTIONS EMPLOYED<sup>(33, 34)</sup>

Solution	A	B	C
Description	Dilute Oxidizing	Concentrated Oxidizing	Concentrated Reducing
Hydrogen Peroxide (g/L)	0.3	1.5	
Acetanilide (g/L)	0.1	0.5	
Hydrazine (g/L)			5
Ethylenediamine	0.15	0.75	3.5
Tetracetic Acid (g/L)			
Sulfamic Acid (g/L)	0.25-0.5	2.5	25
Hexamethylenetetramine (g/L)	0.1	0.5	1
Temperature of Use	250°F-300°F	250°F-300°F	200°F-250°F
pH	1.8 - 2.3	1.5	1.5

At the time of this writing no information was available pertaining to activated crud removal from stainless steel on the bench-scale tests conducted at WAPD.

## 2. Studies Applicable to a Carbon Steel System

For the removal of  $\text{UO}_2$  and fission products from carbon steel, high pH oxidizing solutions, which have been investigated in conjunction with stainless steel, might prove satisfactory. WAPD is presently involved in a study of this problem. The removal of activated crud from a carbon steel system poses a serious problem. The low pH reducing solutions used for removing crud from stainless steels exhibit excessive corrosive attack on carbon steel. Information on decontamination of carbon steel systems is not available.

### C. FULL-SCALE TRIALS ON EQUIPMENT AND PLANTS

Several large-scale runs with reference solution (C) have been carried out in decontaminating stainless steel systems coated with activated crud. Included in this category are the fill-and-flush decontamination of the KAPL-120 loop and the feed-and-bleed decontamination of the WAPD-29 loop.

#### 1. KAPL - 120 Decontamination

In order to proof test the fill-and-flush method for removal of anticipated S3G/S4G corrosion products, decontamination of KAPL-120 loop at WAPD was prescribed<sup>(35)</sup>. This method was chosen because it required the least amount of system modifications for decontamination of the S3G/S4G steam generators.

#### Loop Parameters:

Pumps	3 Westinghouse Type 150C (3 in parallel - 2 in operation)
Nominal loop flow rate	31-33 gpm
Loop Material	SS Type 347
Total loop volume	40 gal. circulating 35 gal. pressurizer
Loop surface area	142 ft <sup>2</sup>
Charging pump capacity	10 and 15 gph

## 2. Test Results:

During this test the in-pile tube had been isolated from the out-of-pile portion of the loop. Composition of the concentrated reducing solution used for this test is given in Table XXXI (solution C). The decontamination factor after a single 3-1/2 hour treatment of the loop followed by repeated water flushes ranged from 2 to 2.5 as evidenced by dose-rate readings at selected points throughout the loop. Samples of the decontamination solution and of the initial water flush indicated that approximately 100 grams of corrosion products ( $\text{Fe}_3\text{O}_4$ ) were removed from the loop surfaces. Radiochemical analyses of the solution established that  $\text{Co}^{60}$  was the major constituent contributing to the gamma dose rate.

At the termination of the decontamination test a 2200 psi hydrostatic test indicated no leaks. Table XXXII lists decontamination factors at various points throughout the loop.

### D. WAPD-29 DECONTAMINATION

The rupture of defected PWR fuel test specimens on June 18 and 26, 1957, resulted in the release of irradiated  $\text{UO}_2$  and possibly  $\text{Al}_2\text{O}_3\text{-UO}_2$  sintered fuel material into the loop resulting in contamination of the facility. After shutdown of the MTR on July 14, 1957, and following water flushings and drainage of the WAPD-29 facility, the radiation fields in the vicinity of the loop prevented carrying out normal MTR shutdown operations. To remedy this situation, chemical decontamination was carried out<sup>(34)</sup>.

A feed-and-bleed decontaminating procedure was employed in this operation. An oxidizing-complexing solution was used to remove  $\text{UO}_2$  and associated fission products. This was followed with a reducing-complexing solution



to remove deposited corrosion products (solutions B and C respectively).

Treatment with the oxidizing-complexing solution yielded an average decontamination factor in the range of 10 to 15. The reducing-complexing solution treatment yielded an additional factor of about 3 to 4 resulting in an over-all decontamination factor of about 50.

Following decontamination, sections of the loop (304 and 347 SS) piping were removed along with six representative welds and subjected to metallographic examination. Emphasis was placed on observing any effects of pitting, cracking, or intergranular corrosion in the weld areas. No deleterious effects resulting from the decontamination operation were detected.

**Loop Parameters:**

Pumps	Two Centrifugal 30-A (One normal, one standby)
Loop Volume	10 Gal., circulating 5 Gal., surge tank
Out of pile area	53 ft <sup>2</sup>
Loop Material	304 SS

**a. Phase I - Decontamination With Oxidizing - Complexing Solution**

Solution (B) (Table XXXI) was utilized for this phase of the decontamination test. The procedure used was of the feed-and-bleed type and consisted of a 4-step sequence as follows:

Step 1: A 2.5 hour treatment with solution (B) containing in addition  
0.2 N HNO<sub>3</sub>.

Step 2: A 2 hour treatment with the solution (B) containing in addition  
0.4 N HNO<sub>3</sub>.

Step 3: A 5.5 hour treatment with solution (B)

Step 4: Circulation of 2 N  $\text{HNO}_3$ - 1 v/o  $\text{H}_2\text{O}_2$  through the loop for 20 minutes (not including time to fill and drain).

Results of this phase of the test are presented in reference 34.

For purposes of this survey it is sufficient to state that decontamination factors varied from 7 to 25 with one extremely high factor of 250. The latter factor was realized in the general subpile room field which at the start of the test exhibited an extremely high radiation level (20 R).

b. Phase II - Decontamination With Reducing-Complexing Solutions

The second phase of the WAPD-29 decontamination was initiated approximately ten days after termination of Phase I. The solution employed in this treatment (solution C) facilitated removal of deposited corrosion product activities along with those fission products sorbed on the crud surface.

The loop was filled with the decontamination solution and brought to a temperature of  $240^{\circ}\text{F}$  at 500 psi over a two hour period. The feed-and-bleed procedure was initiated (bleed rate 0.1 gpm), with the solution circulating through the loop at a rate of 7.5 gpm. After 8 hours of this, the solution in the loop was replaced with water and flushing was carried out for 7 hours. Results of Phase II decontamination test are presented in Table XXXIII.

**TABLE XXXII DECONTAMINATION FACTORS REALIZED DURING  
DECONTAMINATION OF KAPL - 120 LOOP (35)**

Survey Points	Pump #1		Pump #2		Pump #3		Strainer	Heat Ex- changer	Isolation Valve to Purifica- tion Sys.	Average
	Inlet	Outlet	Inlet	Outlet	Inlet	Outlet				
First Decontamination										
A. After drainage of decontamination solution	1.26	1.19	1.62	1.25	2.22	1.35	1.35	2.0	1.37	1.51
B. After one water flush	1.31	1.34	1.66	1.61	2.22	1.81	1.61	2.18	2.14	1.76
C. After five water flushes	1.46	1.88	2.0	2.0	2.95	2.11	1.80	2.5	2.4	2.12
Second Decontamination										
D. After decontamination and two water flushes	1.25	1.63	1.47	2.27	1.42	1.80	1.66	1.15	1.28	1.55
E. Overall decontami- nation (combined effect)	2.17	2.90	3.52	4.54	4.21	3.80	2.50	3.45	3.33	3.38

**TABLE XXXIII WAPD-29 DECONTAMINATION TEST,  
PHASE II, REMOVAL OF DEPOSITED  
CORROSION PRODUCT ACTIVITY (34)**

Date Time	(mr/hr) Loop Radiation Monitor Measurements						Decontamination Factor
	7-26 1700	7-26 1900	7-26 2400	7-27 0100	7-27 0200	7-27 1000	
Loop Cooler	2000	700		350	350	200	10
Circulating Pump	1000	1000	600	700	700	350	3
Max. Pipe Field	1500 to 2500	800	400	400	400	200	7.5 to 12.5
Cubicle Field		300	200	200	200	100	3
Gen. Loop Field	400					75-100	4
Surge Tank (bottom)	1500	400		350	350	200	7.5
Surge Tank (top)	400	200		180	200	70	5.7

## VIII CONCLUSIONS AND RECOMMENDATIONS

As a result of the literature survey and personal contacts, the following conclusions can be made:

1. Large amounts of radioactivity have built up on primary system components at the S-1-W.
2. For stainless steel clad fuel elements, the build-up is expected to be significantly greater than that found on the S-1-W.
3. The activity build-up appears to result from a deposition and/or atom exchange phenomenon.
4. The experimental data available from other operating pressurized water reactors is inconclusive.
5. Since no other operating pressurized water reactor employs stainless steel clad fuel elements, the existing data can only be used qualitatively for the APPR.
6. Theoretical calculations to enable predictions for long-lived nuclides of the total activity build-up have not correlated well with experimental values.
7. Decontaminating chemical solutions and the methods used for applying these solutions are complex. Decontamination factors that have been achieved with these solutions may not be satisfactory in some cases.

An experimental program is necessary to determine the following:

1. How much activity will be present after various intervals of operation?

2. What problems will arise in the repair or maintenance of the reactor system as a result of this build-up?
3. What can be accomplished to reduce this build-up if it should appear that its magnitude is too great?
4. What basic phenomena are occurring to cause this build-up?
5. At what intervals will it be necessary to decontaminate the system should this procedure be necessary?
6. How does one accomplish this decontamination and at what cost?

For the APPR, the experimental program should involve the following:

1. Analysis of APPR primary water and crud.
2. Analysis of test coupons inserted in various locations of the APPR.
3. Measurements of dose rates of the primary system after shutdown.
4. Measurements of dose rates of demineralizer during operation and after shutdown.
5. Effect of such variables as flow rates, water conditions, power level, temperature, materials of construction, location, etc. on activity build-up.
6. Development of suitable decontaminating agents.
7. Development of methods for performing decontamination.

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