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AF NETF
Primary Coolant Conductivity Study

by

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Wright-Patterson Air Force Base, Ohio

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TECHNICAL REPORT

AFIT TR 67-9

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AF NETF Primary Coolant Conductivity Study Abstract

The AF NETF, a 10 MW water-cooled research reactor, monitors the conductivity of its coolant to detect erosion/corrosion of its structural systems and fuel element failure. When a gradual increase in this conductivity sent it above the defined limit, a study was initiated to identify the contaminants and examine the source and removal processes. The major contaminant, Na^{23}, was traced to an in-leakage from the secondary to the primary coolant systems through the heat exchanger interface. Minor amounts of contaminant were introduced from a deluge tank and from residual ions remaining on the demineralizer from the factory NaOH flush. Second order contributions stemmed from: (a) fission fragment migration through the fuel element cladding--directly proportional to the cumulative hours of reactor operation and the fuel-coolant temperature gradient and (b) erosion/corrosion of the fuel-element cladding and reactor structural and piping materials--directly proportional to the coolant temperature and flow rate. Clean-up was found to be directly proportional to the flow rate through the demineralizers, up to the 60 gpm pumping limit of the cleanup loop with a diminishing effectiveness past the point of resin channeling--an estimated 50 gpm/ft^2 of resin bed.

The study proved the absence of any fuel cladding defect and the presence of only an acceptably minimal structural erosion/corrosion rate. The in-leakage source was identified and corrected. The demineralizer system was found to be adequate for normal operation, but economically restrictive during the latter portion of resin life. A conductivity specification of 3 μmho/cm was found to provide an engineeringly-sound definition of increased erosion/corrosion or cladding failures and to be in keeping with specifications defined at comparable facilities. This economically desirable and engineeringly acceptable specification is recommended for this facility.

The views expressed herein are those of the author and do not necessarily reflect the views of the United States Air Force or the Department of Defense.

AF NETF Primary Coolant Conductivity Study

I. INTRODUCTION:

The Air Force Nuclear Engineering Test Facility at Wright-Patterson Air Force Base, Ohio, is a 10 MW water-moderated and water-cooled unpressurized research reactor. Conductivity of the reactor primary coolant media is monitored to provide a gauge of system erosion/corrosion rates and operating limits are defined in the governing set of Technical Specifications. When this monitored parameter exhibited a constantly increasing value and the conductivity levels approached and surpassed the recommended operating limits, a study was initiated that would define the factors contributing and corrective actions required. The study was to encompass an engineering evaluation and solution only. Any additional detailed analyses of contributing factors would be left for subsequent experimental studies.

The engineering study was a two-part effort that first examined the contaminants contributing to the impurity and their associated growth rates and then examined possible reactor operating parameters that might account for these growths. The first part of the study would identify any erosion/corrosion of reactor components and expose any fuel element rupture with accompanying fission product release. The second part of the study would examine a time-rate-of-change of primary coolant conductivity as pertinent reactor operating parameters were varied and would ultimately define corrective actions required.

II. CHEMICAL/RADIOCHEMICAL ANALYSES:

Exhaustive chemical and radiochemical analyses of the primary coolant waters were accomplished by the Taft Laboratories of the United States Public Health Service, Cincinnati, Ohio. The samples analyzed were accurately identified as to reactor operating status and history and were analyzed as quickly as possible after sampling to capture the effects of short-lived isotopes. These analyses conclusively proved the absence of fuel element cladding rupture and verified a very minimal structural corrosion rate. The nuclides expected as daughter products of U^{235} fission were not present. The Al, Ag, Mg, Be, and Cd ions expected from the erosion of the aluminum clad fuel elements, beryllium reflectors, and cadmium control rods were present in only infinitesimal amounts. (See Tables 1 and 2).

TABLE 1

Stable Ions in Reactor Coolant

Ion	mg/l			Method of Analysis
	Primary Inlet	Cation Outlet	Mixed Bed Outlet	
Cr^{+3}	0.0005	< 0.0003	< 0.0003	a
Mo^{+3}	0.0004	0.0001	< 0.0001	a
Fe^{+3}	< 0.004	0.004	0.008	a
Na^{+}	0.15	0.15	0.2	b
Cl^{-}	< 0.17	< 0.17	< 0.17	c
SO_4^{-2}	< 0.2	< 0.2	< 0.2	c

a Emission Spectroscopy
b Atomic Absorption
c Wet Chemistry

Tables 1 and 2 reflect the result of a monitored test while the reactor was operated at 10 MW from 0022/22 Aug - 1742/30 Aug 66. Samples of the reactor coolant were collected on 24 Aug, 29 Aug, and 1 Sep 66 for analysis by the Taft Sanitary Engineering Center, USPHS, Cincinnati, Ohio. Their report, dated 10 Oct 66, stated in part:

"The preliminary analyses on the samples of reactor coolant collected August 24, August 29, and September 1 have been completed. Each of the samples was gamma scanned repeatedly at a specified geometry to identify photopeaks by decay measurements. At the same time, chemical separations were performed primarily on the primary inlet, and stable ions were measured either by atomic absorption, emission spectroscopy, or wet chemistry. The results are listed in Tables 1 and 2..."

TABLE 2

Radionuclides Identified in Reactor Coolant

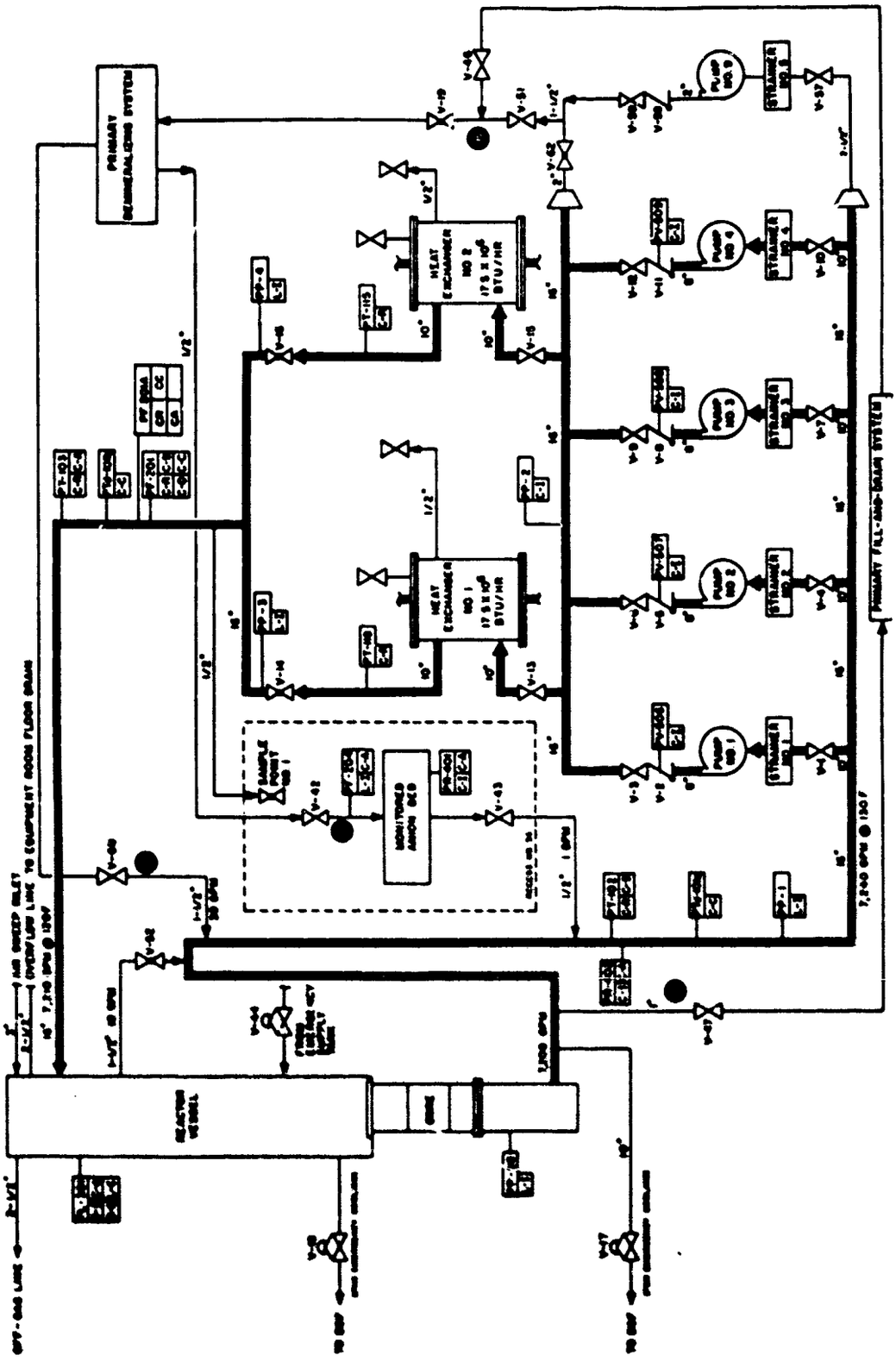
Nuclide	t 1/2	Principal γ Photopeak	μCi/ml	Remarks	Sample Source
^{24}Na	15 h	1.368	2.1×10^{-2}	Activation Product	Primary Inlet 8/29
^{115}Cd	2.3 d	0.52	5.8×10^{-5}	"	"
^{99}Mo	2.8 d	0.142, 0.75	6.6×10^{-5}	"	"
^{32}P	14 d	β	4.3×10^{-6}	"	"
^{55}Fe	2.6 y	X-ray	2.5×10^{-7}	"	"
^{60}Co	5.3 y	1.173, 1.33	1.0×10^{-7}	"	"
^{54}Mn	300 d	0.84	1.7×10^{-7}	"	"
^{110m}Ag	253 d	0.66, 0.89, 1.38	3.1×10^{-7}	"	"
^{3}H	12 y	β	6.0×10^{-4}	"	"
^{65}Zn	245 d	1.119, 0.51	$< 1 \times 10^{-7}$	"	"
^{51}Cr	28 d	0.325	2.3×10^{-5}	"	Primary Cation Outlet 8/29
^{124}Sb	60 d	0.603, 1.69	4.0×10^{-7}	"	"
^{41}A	1.8 h	1.3	3×10^{-4}	"	Mixed Bed Outlet 8/29
^{133}I	21 h	0.53	2.6×10^{-5}	Fission Product	Primary Inlet 8/29
^{131}I	8.08 d	0.364	1.4×10^{-6}	"	"
^{91}Sr	9.7 h	0.551	3.2×10^{-5}	"	"
^{89}Sr	50 d	β	2.0×10^{-7}	"	"
^{140}Ba	12.8 d	0.16, 0.54	1.4×10^{-6}	"	"

Nuclide	t 1/2	Principal Photopeak	Ci/ml	Remarks	Sample Source
^{133}Xe	5.3 d	0.081	1.4×10^{-6}	Fission Product	Mixed Bed Outlet 8/29
^{135}Xe	9.1 h	0.25	2×10^{-5}	"	Primary Cation Outlet 8/29
Anion (?)	short	0.52	-1	"	Primary Inlet 8/24
Cation (?)	12 h	0.25	10^{-4} (?)	"	Mixed Bed Outlet 8/29

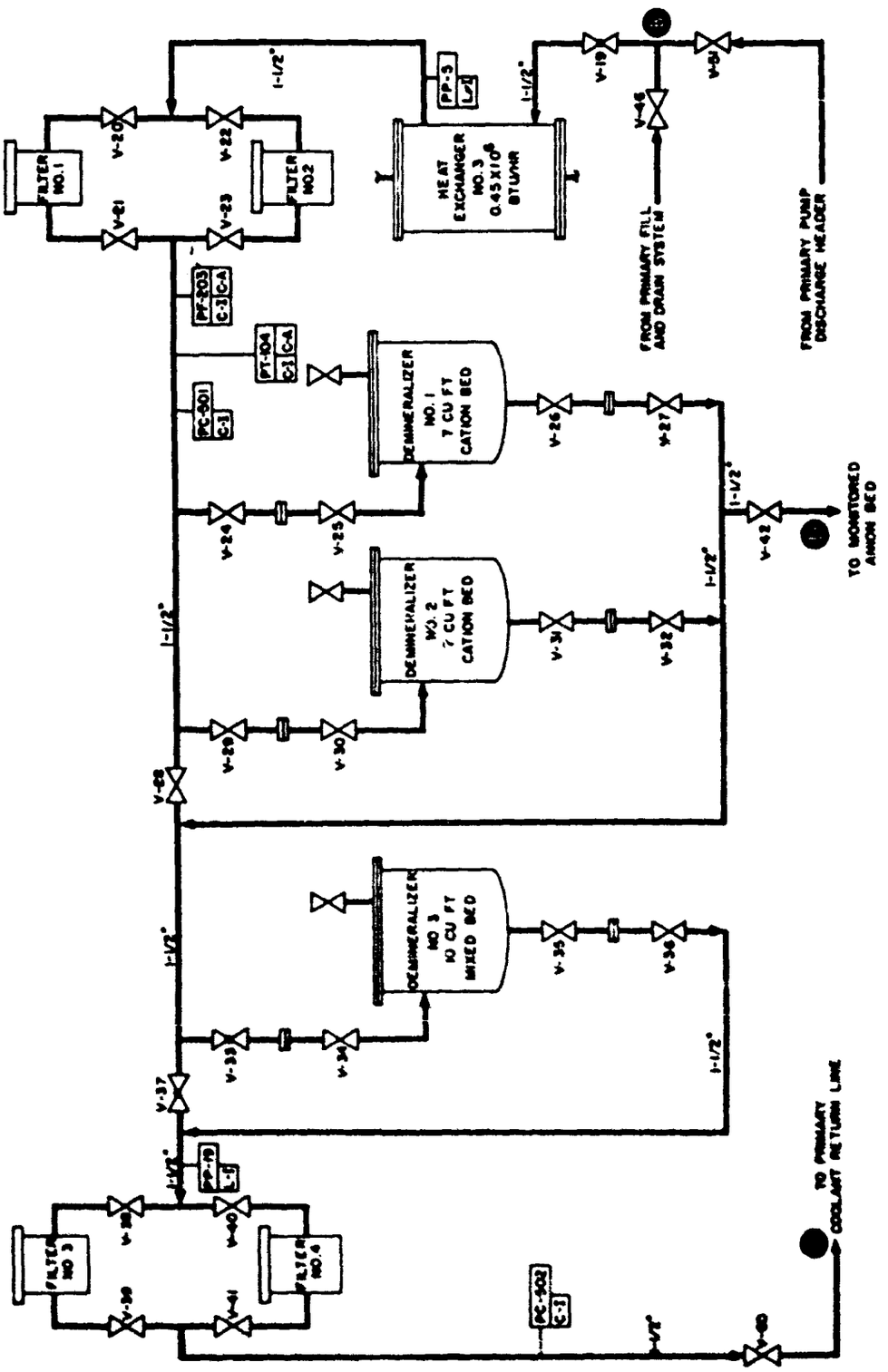
Minor amounts of Na^{24}, attributed to neutron activation of the aluminum fuel-element cladding, were identified, but stable Na^{23} was the dominant ion and this was present in sufficient concentration to account for the conductivity levels observed.

The operation of the total composite system was analyzed to identify possible external source(s) of contaminant. The secondary coolant loop was suspected with a possibility of in-leakage through the counter-flow heat exchanger interface. (See Figures 1 - 3). Although this was ultimately found to be the major source of contaminant, the absence of other trace elements associated with the "softened" secondary coolant waters initially negated this as a suspect. The pipes and valves connecting the primary system to a 95,000 gallon Na_2CO_3 - stabilized deluge tank were examined and a surge line was installed to eliminate any minor in-leakage. The primary system was a closed loop and all make-up waters were first demineralized so this could be, at worst, a minor contribution. The one remaining source of contaminant introduction, the demineralizer resins themselves, was analyzed and a large amount of free Na^+ ion was found as remaining from the factory NaOH flush. The demineralizer resins and all in-line filters were replaced with specially procured sodium-free resins and clean filters to eliminate this as a suspect area and to provide a clean system from which to observe contaminant buildup.

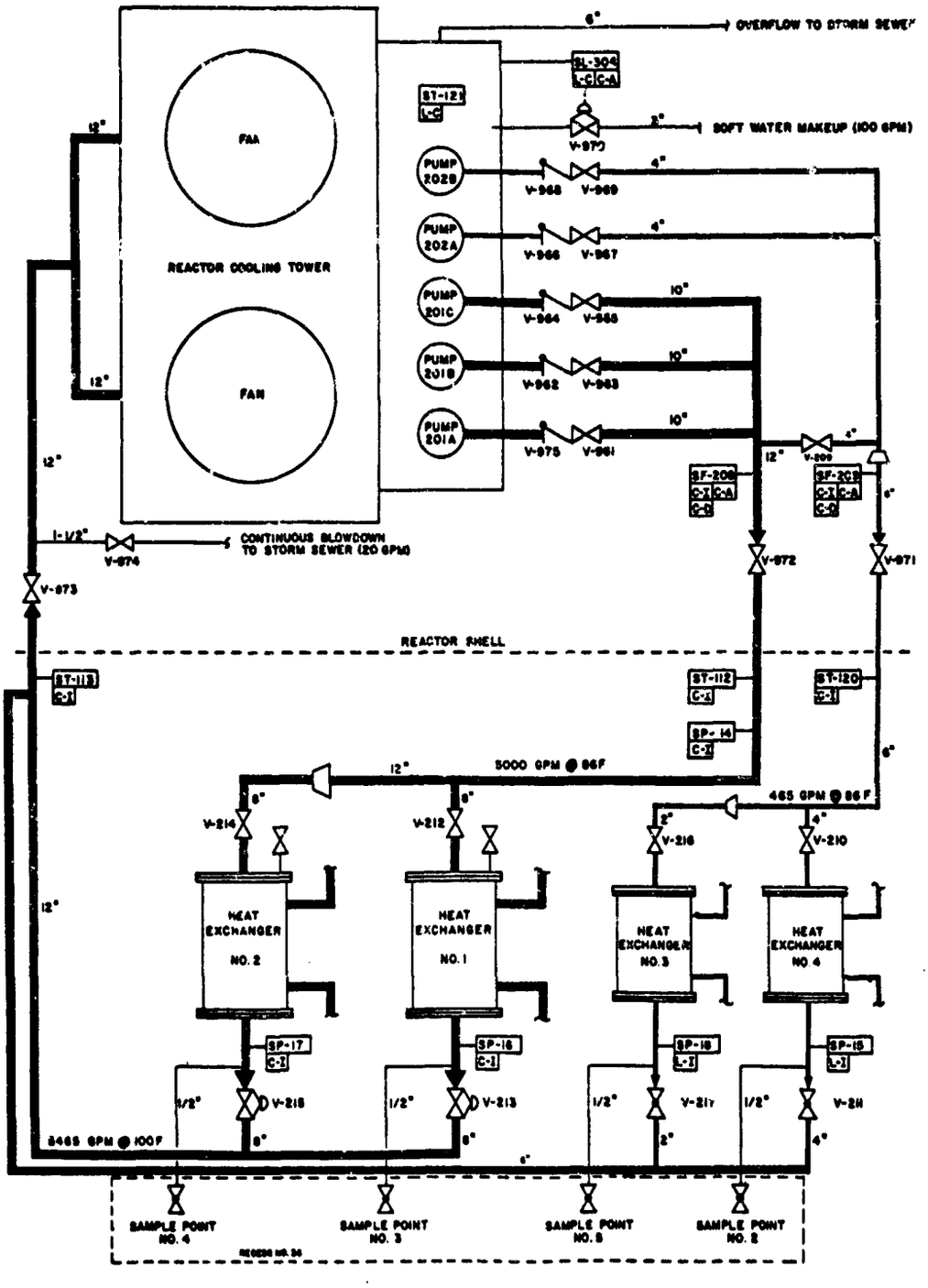
In summary, initial chemical/radiochemical analyses showed that sodium ions accounted for the major portion of the contaminant with stable Na^{23} by far the major contributor. It had been postulated that the monovalent Na ions could be collected by the cation resin beds during reactor shutdown, but that they could subsequently be displaced by the bivalent and trivalent fission and erosion products to account for the increased activity observed during reactor operation. With this assumption, an increased fission fragment inventory resulting from cumulative hours of reactor operation could provide an increased migration gradient through the fuel element cladding, increased fuel element temperature gradients with increased reactor power could enhance fission fragment release and cladding corrosion, and increased primary coolant temperatures and flow rates could result in increased erosion/corrosion rates. If the assumption was not valid, or insignificant in effect, the contaminant could only stem from an initial introduction from an as yet unidentified external source. The answer could be found only through a closely controlled testing program.



PRIMARY COOLING SYSTEM



PRIMARY DEMINERALIZING SYSTEM



SECONDARY COOLING SYSTEM

III. REACTOR PARAMETRIC STUDY:

An examination of the parameters possibly contributing to the primary coolant conductivity showed it to be a net result of opposing efforts. The study showed the contaminant-producing source processes to be opposed by demineralizer cleanup processes and the accuracy of measurement of the net result to be inhibited by temperature effects on the conductivity probes. Further, the several source and removal processes were, in themselves, interrelated and not clearly separable. Transient changes in conductivity following controlled changes in parametric settings caused observations of less than 24 hours to be fallacious and often misleading. In short, a series of parameters were varied in a controlled pattern, but resultant effects were often so minor and so gradual as to give misleading indications and an extensive program was required before a correct solution was achieved.

The contaminant production rate, or source rate, was postulated to be a function of (1) reactor power level and cumulative hours of reactor operation with the associated increase in fission product contaminant caused by diffusion into the coolant, (2) temperature, ion content, pH, and flow rate of the primary coolant with their associated effects on erosion/corrosion rates, and (3) external contaminant sources such as in-leakage and residual ions on supplied resins. The cleanup rate was postulated as a function of (1) demineralizer volumes and resin types, (2) flow rate through the demineralizer beds with increased flow resulting in a greater fraction of the total coolant loop being processed, but at a cost of a decreased stay time within the demineralizers, and (3) temperatures within the demineralizer beds. The conductivity probes were known to be minutely temperature sensitive so, despite calibration and compensation efforts, the apparent changes in conductivity resulting from controlled changes in operating parameters were suspect to a minor degree. The portion of reactor engineering dealing with corrosion of cladding and structural materials and that facet of water chemistry dealing with demineralizer ion exchange columns remains partially within the frontier of the technology. The study became a systematic separation of contributing parameters and then a close analysis of conductivity levels as those parameters were individually changed. The study would identify and eliminate or minimize source terms, evaluate and optimize cleanup processes, or provide an engineeringly sound redefinition of the conductivity operating limit if such were indicated. Preliminary evidence indi-

cated that in-leakage sources had been eliminated, that contaminant ions were no longer introduced from the demineralizer resins, and that the source term was now related to fission products migrating through the fuel element cladding and erosion products from the reactor structural and piping systems. Experimentation would corroborate or disprove this evidence and examine methods of improving the cleanup processes.

The first parameter examined was the demineralizer flow rate. This had been maintained at a nominal 30 gpm and would now be examined at incremental steps from some minimal amount to the maximum attainable. This represented the first physical testing accomplished and the interplay between operating parameters was exposed as testing progressed. Testing experience also demonstrated the transient effects accompanying a change in parametric setting and the requirement for waiting for system stability before evaluating results. The experiment ultimately evolved into a series of 16-hour runs at the various flow rates with a minimum of 10 hours required for system stabilization and a minimum of 6 hours for a reliable definition of trend. The reactor power level, the reactor coolant loop temperature, and the demineralizer coolant loop temperature was maintained constant throughout the test. Changes in ambient temperatures made close control of loop temperatures difficult and changes of 1 - 2°F in the coolant temperatures were reflected in perturbations of the graphic data. The parametric effects and control requirements were identified only through experience and changes in testing procedures were required. The data was plotted as a time-rate-of-change of conductivity with a zero or negative slope desired. While this optimum was not achieved during this portion of the test, the results showed continuing improvement with increasing flow up to 60 gpm, the limit of the pumping system. The manufacturer quoted an upper limit of 50 gpm/square foot of demineralizer bed to preclude resin channeling. Design studies were made to permit increased demineralizer flow should it be required.

Reactor coolant loop temperatures and reactor power levels were varied independently with all other parameters held constant. The time-rate-of-change of conductivity was shown to be directly proportional to the reactor power level and directly proportional to the coolant temperature. The system was run with the reactor shut down and at various power levels, with various demineralizer flow rates, and with various primary coolant loop temperatures. This series of controlled combinations showed that nuclear operation contributed to the increase in conductivity, but that the major increase occurred

independent of reactor operation in direct proportion to the operating temperature.

A re-evaluation of the secondary coolant relationship was conducted. Control of the primary coolant temperature is partially achieved by throttling the secondary coolant flow through the tubes of the tube-and-shell heat exchanger. This throttling increases the pressure within the secondary system and, since the primary system is maintained at a negative pressure with respect to the secondary to insure in-leakage in the event of failure, an increased secondary-primary differential pressure is established. A meticulous chemical testing program conducted simultaneously with an exaggerated overpressurization revealed an in-leakage of the secondary coolant into the primary system. The sodium ions associated with the water softeners used with the secondary coolants were the contaminants contributing to the primary coolant conductivity. The leak was isolated and repaired and the conductivity of the system immediately improved. A stable conductivity level of better than 0.3 μ mho/cm was maintained throughout the following extended reactor runs with a demineralizer flow rate of 30 gpm and a nominal 105°F average reactor coolant loop temperature.

A rather abrupt decline in conductivity with an associated increase in pH near the end of the run signaled the impending depletion of the mixed-bed demineralizer. This was followed a short time later by a failure of first one and then the other of the cation beds. These resins were replaced to immediately restore the system to proper operation. It would appear that the abnormal requirement imposed by the secondary in-leakage had prematurely depleted these resins and that a longer life might be expected from subsequent charges. Subsequent operation will now define the resin-bed lifetime to be expected from normal operation.

IV. SUMMARY:

An extensive and exacting testing program was conducted to determine the reason for change in the primary coolant conductivity value and to provide a means of returning it to the operating limit or an engineeringly sound redefinition of the limit. The tests showed that the contaminants contributing to an increased conductivity do actually stem from fission-induced sources and from erosion/corrosion processes--the actual reason for monitoring the parameter--but that they may also be introduced from external sources. The major source of contaminant(s) were, in fact, from these external sources and they

were extremely difficult to define because of their minuteness and parametric interplay. It was determined that the time-rate-of-change of conductivity was directly proportional to the reactor power level and primary coolant temperature and inversely proportional to the rate of flow through the demineralizer up to the channeling limit.

This study is terminated with the identification and correction of the problem causing the improper conductivity level. As indicated in the Introduction, it is not within the scope of the study, or the operational program of this facility, to pursue the interesting follow-on topics that evolved. What are the mechanics involved in fission product migration and what is the effect of varying the fuel/cladding/coolant temperature gradient? What is the detailed mechanism of ion removal by demineralizer resins and how prevalent is the replacement of monovalent ions by bivalent and trivalent ions? How is apparent conductivity affected by the temperature of the media and ion mobility? What are the exact contributions of each of the defined parameters and how might the system or operating conditions be modified to provide a greatest possible water purity? These questions raised during the course of this program, though worthy of examination, must be referred to a subsequent study.

The demineralizer system within the AF NETF was found to be adequate to maintain the system conductivity within the prescribed operating limit, except when the demineralizer resins neared the limit of their operating life. Studies showed comparable reactor systems to employ a conductivity specification of 2 - 3 μ mho/cm as opposed to the 1 μ mho/cm employed at this facility. This relaxed specification was shown to be engineeringly sound in that the erosion or cladding failures that this parameter monitors will result in clearly definable changes in conductivity values and would indicate the adoption of that specification at the AF NETF.

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13. ABSTRACT

The AF NETF, a 10 MW water-cooled research reactor, monitors the conductivity of its coolant to detect erosion/corrosion of its structural systems and fuel element failure. When a gradual increase in this conductivity sent it above the defined limit, a study was initiated to identify the contaminants and examine the source and removal processes. The major contaminant, Na^{23}, was traced to an in-leakage from the secondary to the primary coolant systems through the heat exchanger interface. Minor amounts of contaminant were introduced from a deluge tank and from residual ions remaining on the demineralizer from the factory NaOH flush. Second order contributions stemmed from: (a) fission fragment migration through the fuel element cladding - - directly proportional to the cumulative hours of reactor operation and the fuel-coolant temperature gradient and (b) erosion/corrosion of the fuel-element cladding and reactor structural and piping materials--directly proportional to the coolant temperature and flow rate. Clean-up was found to be directly proportional to the flow rate through the demineralizers, up to the 60 gpm pumping limit of the cleanup loop with a diminishing effectiveness past the point of resin channeling--an estimated 50 gpm/ft^2 of resin bed.

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