Atomic Energy of Canada Limited

FOULING IN ORGANIC-COOLED SYSTEMS

CRCE-1096

by

D. H. CHARLESWORTH

Chalk River, Ontario
April, 1963

AECL-1761

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SYNOPSIS

Studies of organic coolants in the out-reactor 250-0-1 loop and in the in-reactor X-7 loop have shown that fouling films are deposited on heat-transfer surfaces by two mechanisms, one involving soluble impurities and the other insoluble impurities in the coolant. The simultaneous action of two mechanisms of deposition can lead to a wide variety of compositions and structures of the deposited film.

The concentration of impurities is the most important factor controlling the deposition rate. Coolant velocity and surface temperature also have major effects on the fouling rate. At low chlorine levels continuous coolant cleanup through Attapulgus clay has been shown to reduce deposition rates under representative reactor conditions from 100 $\mu g/cm^2$ hr to 1 $\mu g/cm^2$ hr. Chlorine, which is a strong promoter of fouling, is not removed by Attapulgus clay. Further studies of its important effect on fouling and its removal will be the subjects of separate reports.

An acceptable deposition rate of 0.3 $\mu g/cm^2$ hr should be achieved by intensive purification, coupled with the exclusion of impurities such as chlorine.

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1. INTRODUCTION

Atomic Energy of Canada is at present engaged in the development of organic liquids as reactor coolants. The coolant of immediate interest is a mixture of terphenyl isomers plus the associated decomposition products which are held at an equilibrium concentration of about 30 wt % by a purification side-stream. Since there is an economic incentive to design for sheath temperatures as high as permitted by the SAP fuel sheathing there has been concern about the possible deposition of material on fuel element heat transfer surfaces. Any such deposit or fouling could have detrimental effects on fuel performance through two mechanisms both of which result in higher fuel temperature. The deposit could present an added resistance to heat transfer from the fuel to the coolant. Also, the deposits might reduce the coolant flow around the fuel elements either by roughening the elements or by actual restriction of the coolant channel. In a reactor, the coolant flow passages over the fuel may be as narrow as 0.1 cm (0.040 in.), and the heat fluxes could be in excess of 100 watts/cm^2 (317,000 BTU/hr ft^2). The limit on the fuel is usually its surface temperature because of material problems. Hence, an increase in surface temperature of 25°C (45°F) above the design value is about all that can be tolerated. A fouling film only 25 μm (.001 in.) thick could give such an increase. Therefore, the rate of fouling must be less than 25 μm (0.001 in) per year over the fuel life of one or two years.

Early experience in the United States using polyphenyl coolants in test loops both in-reactor and out-reactor was encouraging from the fouling point of view. Loop operation by Atomics International (1) in the MTR and by Monsanto (2) in the Brookhaven Reactor showed no significant fouling. The Organic-Moderated Reactor Experiment (OMRE) was started up in February 1958 and operated successfully with its first core. In out-reactor loop studies at AI using coolant from OMRE the measured rates, although higher than desirable, were in a practical range for operation. However, in the latter part of 1959 during operating of the OMRE with its second core increasing difficulties with fouling were encountered although operating temperatures and flows were not greatly different from those in Core I operation (3).

Parkins (4) and Gercke (5) of AI have analysed the observations of fouling in the OMRE and test loops and concluded that a single basic process dependent upon the presence of suspended solids in the coolant had been responsible for the production of surface films. Iron was assumed to have been introduced into the coolant as corrosion products from the mild steel system. The corrosion was accelerated from air and water which entered the coolant during shut-down operations. The process, as proposed by Parkins, involved transport of these particles to the surface and attachment there through the establishment of chemical bonds. Brownian motion appeared to be the principal mechanism which brought the particles in contact with the surface but many factors could have been important in determining whether a given encounter lead to permanent attachment of the particle. Within a reactor core, the nuclear radiation field can be effective in accelerating the chemical bonding, a function performed only by thermal processes in conventional situations.

A program of fouling studies in organic cooled systems, is being conducted at AECL. This report covers results obtained up to April 1962. Subsequent results particularly on the important effect of the fouling promoter chlorine, will be the subject of a second report to be issued shortly.

2. EXPERIMENTAL STUDIES

Observations and measurements of fouling have been made from tests with organic coolant in the 250-0-1 out-reactor loop and from fuel-element irradiations in the organic-cooled X-7 loop in the NRX reactor. Although most of the irradiation tests were devised for testing fuel elements, fouling information has been produced as an important by-product.

In all cases the coolant was a mixture of ortho- and meta- terphenyls in the ratio of about 2:1 blended with 30 wt % OMRE high boilers from Core I and II. This mixture was purified in various ways both before and after charging to the loops.

2.1 The 250-0-1 loop

The 250-0-1 loop, shown schematically in Figure 1 is a mild steel loop. The coolant is circulated by a

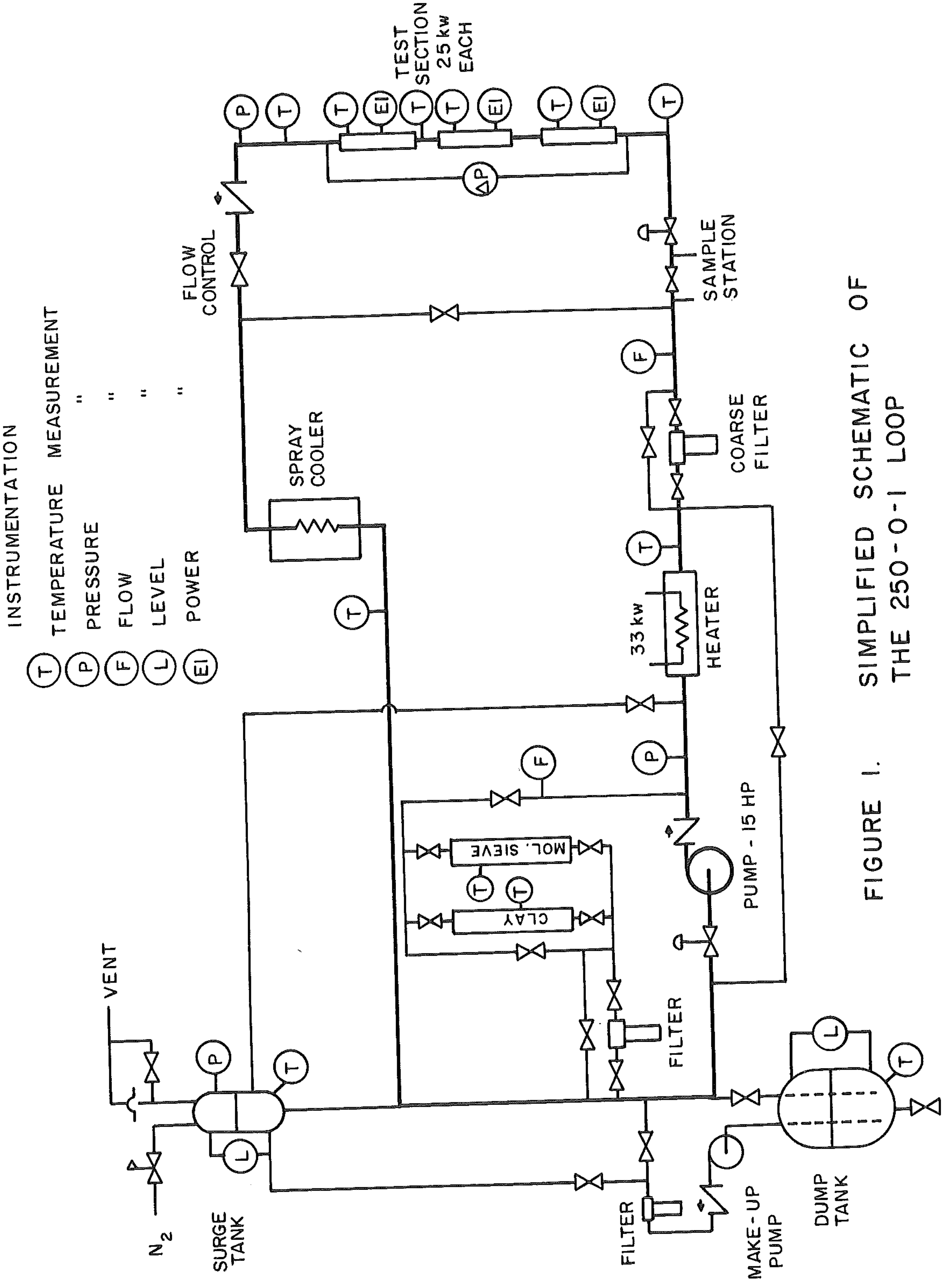


FIGURE 1. SIMPLIFIED SCHEMATIC OF THE 250-O-1 LOOP

mechanically sealed centrifugal pump through a heater, a flow-metering orifice, three test sections in series and a cooler. A side-stream can be passed through a purification circuit. Pressure is applied on the coolant by nitrogen in the surge tank. Normally, no liquid or gas flow is passed through this surge tank.

No extensive cleaning procedure was applied to the loop at the time of construction. Prior to assembly loop components were wire-brushed and blown free of loose scale. The loop was initially charged with hydrogenated terphenyl (Monsanto HB-40) which was circulated for a few days at temperatures up to 315°C with the full flow of approximately 0.7 l/sec through a 200-mesh filter. After this flush the loop was filled with the terphenyl-high boiler mixture. After two-weeks circulation with a flow of 0.1 litres/sec through 20-µm and 5-µm filters in the bypass circuit the iron content of the coolant had been reduced from approximately 60 ppm to 2 ppm.

The test sections shown in Figure 2 are similar to the type used by Atomics International in their out-reactor loop studies (6). Each test section consisted of a thin-walled stainless steel tube about 3 ft long with coolant flow internally; three electrodes were attached to the tube through which low voltage AC power was supplied. Each tube thus had two regions in which the wall was resistance-heated. Chromel-alumel thermocouples were spot-welded at intervals along the outside of the tube. The tube was encased in two safety shrouds separated by thermal insulation. Normally during any test power was supplied to only two test sections. The third test section was used before and after the test as a reference to evaluate changes in the instrumentation and in the coolant heat-transfer properties.

During a test the coolant temperature and power to the test sections were maintained constant. Twice daily measurements of the heat transfer coefficient in the test sections were made. The coolant was routinely sampled for analysis. At the end of the test, the test sections were removed, rinsed in xylene and cut open. The appearance, both macroscopic and microscopic, of the deposits was recorded. Samples were removed for cross-sectioning by metallographic techniques. A deposit was then removed from portions of the test section by scraping and its composition and weight per unit area determined.

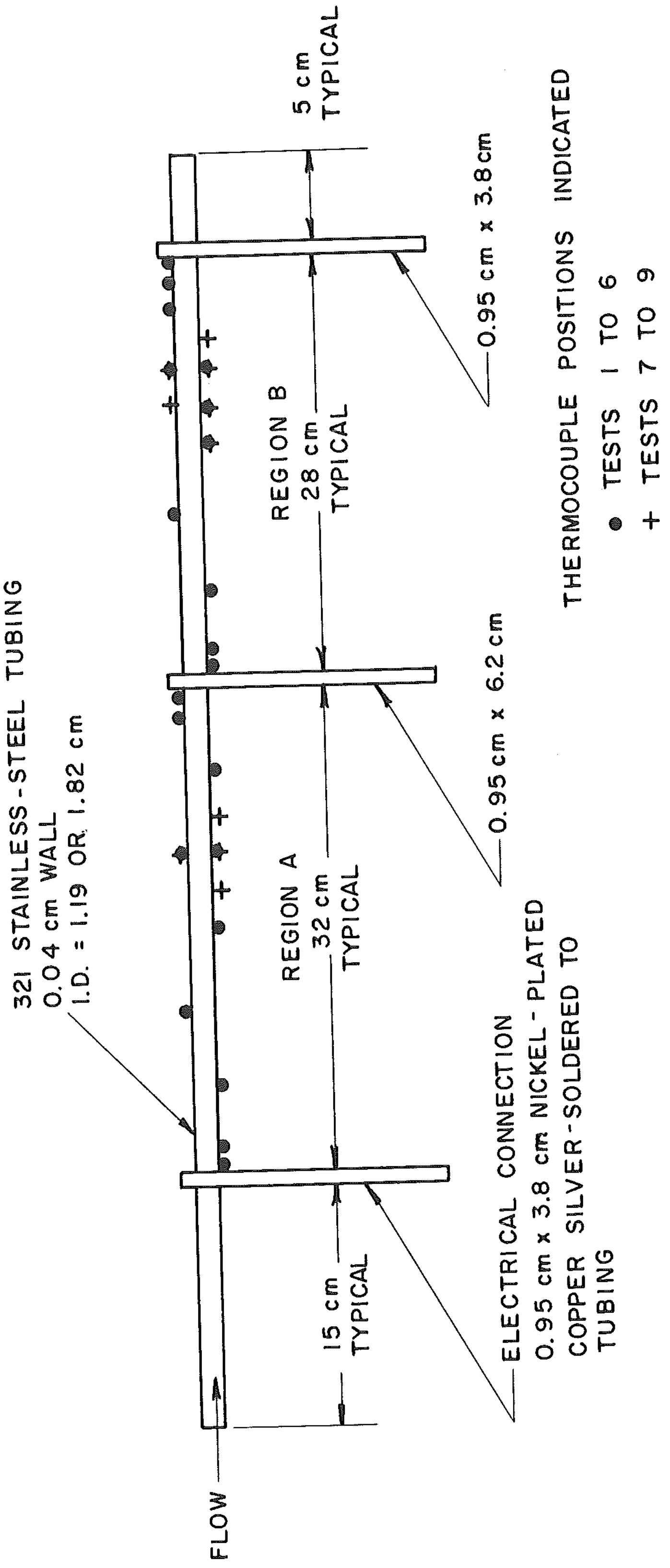


FIGURE 2. SCHEMATIC OF 250-O-I LOOP TEST SECTION

2.2 The X-7 Loop

The X-7 loop shown schematically in Figure 3 is also a mild steel system except for the test section which is of stainless steel. The test section is mounted in the D-22 position of the NRX reactor. The primary circuit includes the pumps, heaters, test section and flow instrumentation. Auxiliary circuits are provided for cooling, purification, surge tank and coolant make-up. Details are listed in Table 1. A wide variety of designs of fuel elements have been irradiated in the X-7 test section. Following a suitable cooling period after irradiation the elements were examined in the Universal and Metallurgy hot cells. Observations of the extent, appearance and thickness of any deposits were determined. In many cases samples of the film were removed by scraping for subsequent analysis.

During the tests, the coolant was routinely sampled and analysed.

3. EXPERIMENTAL RESULTS

3.1 Out-reactor Tests

3.1.1 Out-reactor Fouling Test No. 1

Test No. 1 was mainly for the purpose of testing and becoming familiar with the loop. During the test, the loop and the operating procedures were undergoing frequent modification. The power instrumentation was changed completely part way through the test (138 hours of operation). Corrections have been applied to the data from this early period to make the measured heat transfer coefficients equivalent to those in the later periods. Later in the run, surface temperatures were increased in an attempt to produce fouling of measurable thermal resistance.

Prior to Test No. 1 during loop acceptance tests this coolant was circulated at temperatures mainly between 260° and 370°C but up to 450°C for a period of about one hour. Test No. 1 was started and run for 138 hours with intermittent filtering. A three-week shutdown followed during which additional coolant was added to the system and the whole charge mixed and filtered for one week at 260°C. This brought the iron content of the coolant down to 2 ppm and the test was resumed. At the completion of the test the coolant was drained into the dump tank in preparation for further loop modifications.

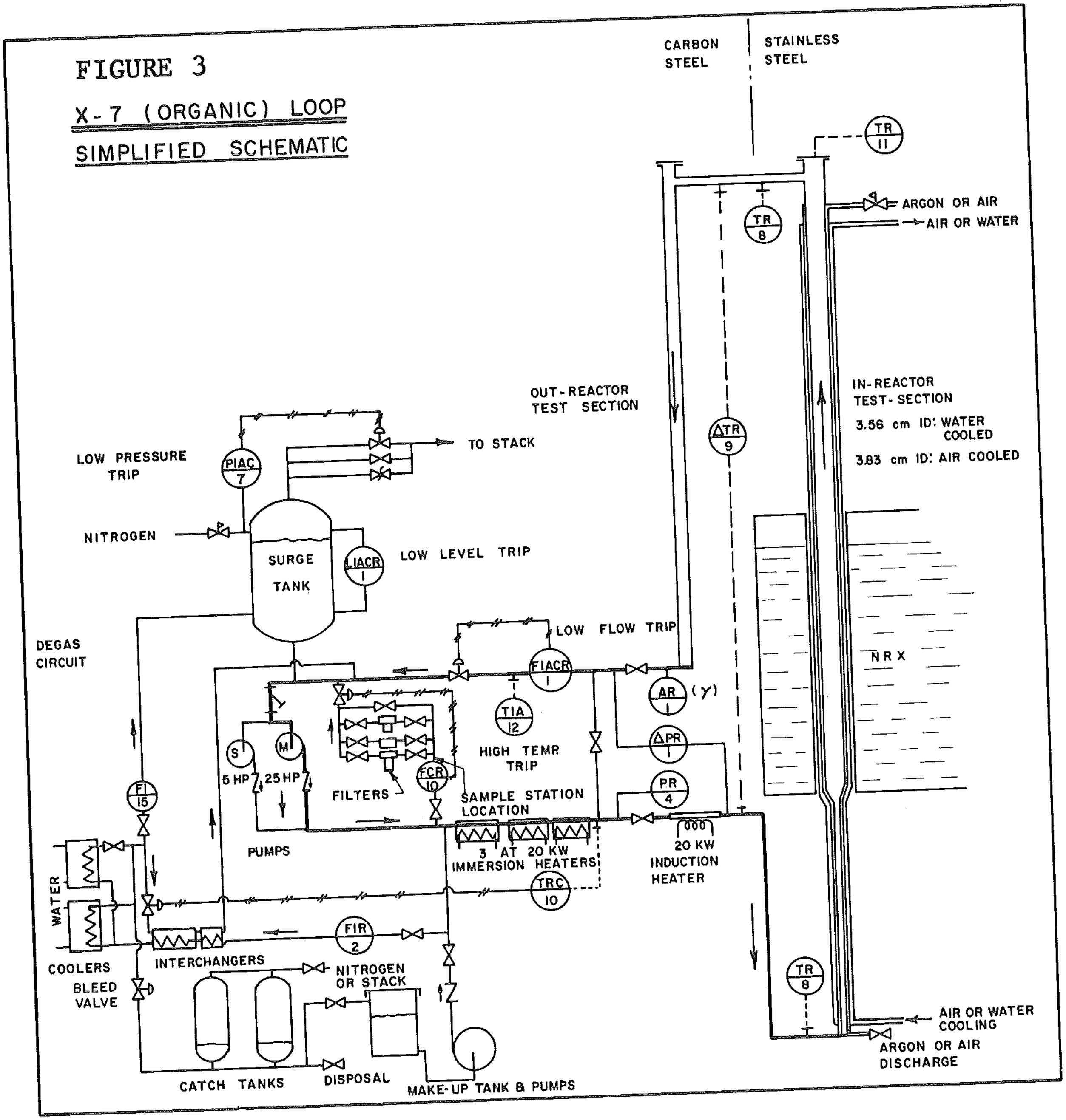


FIGURE 3

X-7 (ORGANIC) LOOP

SIMPLIFIED SCHEMATIC

TABLE 1

22/V/62
R. D. Delaney

AECL X-7 (ORGANIC) LOOP

CAPABILITY: 23 atm at 430°C at pump outlet
Coolant Pumpable at room temperature
3.56 cm ID P.T.; 2 l/s at 3.7 atm across test assembly
1.5 l/s at 6.8 atm " " "
3.83 cm ID P.T.; 2 l/s at 4.7 atm " " "
1.5 l/s at 7.4 atm " " "

SAMPLING FACILITIES:

Hot Liquid Sample Bomb Pyrolytic Fouling Probe	Before adsorption column After adsorption column
Cooled Liquid Sample	Before adsorption column After Adsorption column After filter
Gas Sample Bomb	Surge tank vapour space

REACTOR TRIPS:
Low Flow, Low Pressure
High Temperature, Low Level
Main Pump Off
Standby Pump Not Available
Fire

PRESSURE TUBE:

	Mark I	Mark II
Internal Diameter	3.56 cm 1.402 in.	3.835 cm 1.510 in.
Plug gauge	3.53 cm 1.380 in.	3.83 cm 1.506 in.
Length in flux	305 cm 10 ft	305 cm 10 ft
Volume in flux (empty)	3.0 l 0.80 gal/(US)	3.5 l 0.93 gal/(US)
Material	AISI 321 SS	AISI 321 SS
Heat Loss at 370°C / 700°F	Water Cooled	Air Cooled
over total 10½ m / 35 ft	14 kW	7 kW
over 3 m / 10 ft in flux	4 kW	~ 1 kW

MAIN CIRCUIT:

Pump (Motor)	120 m 400 ft head	(18½ kW) (25 HP)
Heaters: Immersion	3 @ 20 kW	$1\frac{1}{2}$ w/cm^2 (9 w/in.^{2})
Induction	1 @ 20 kW	6 w/cm^2 (40 w/in.^{2})
Orifice	100 in. (H_2O)ΔP ≡ 25 l/s @ 370°C 37 gal(US)/min @ 700°F	
Control Valve	air to close	
Activity Monitor	0-100 to 0-250,000 c/min (gamma)	
Volume	approx. 190 l 50 gal(US) without Induction Heater approx. 230 l 60 gal(US) with Induction Heater	
Heat Loss @ 700°F	50 kW	
Circuit Time @ 15 gal(US)/min	26 s fuel to monitor 156 s fastest 200 s average	

INSTALLATION:
3.56 cm ID Pressure Tube in NRX D-22 (I-1) 25/VII/60 → 4/IV/61
3.83 cm ID Pressure Tube in NRX D-22 (II-1) 4/IV/61 → 27/XII/61
(II-2) 22/I/62 → ....

MEASUREMENTS (RECORDED):
Test Assembly: Temperature
3 Absolute
1 Reference Absolute
18 ΔT or T (adjustable range)
Loop:
Temperature, Temperature Difference,
Pressure, Pressure Difference
Flow, Level, Activity

LOOP TRIPS:
(Full Cooling, No Reactor Trip)
Low Air Pressure to Cooling Jacket
High Temperature in Cooling Jacket

COOLING CIRCUIT:

Heat Removal Capacity	200 kW
Volume	95 l 25 gal(US)
Flow rate (varies automatically to maintain temperature)	0 to 1.9 l/s 0 to 30 gal(US)/min

SAMPLE/FILTER CIRCUIT:

Volume - circulating	60 l 16 gal(US)
- standby equipment	37 l 9.7 gal(US)
Flow rate	12→63 ml/s 0.2→1.0 gal(US)/min
Adsorption Column	
Diameter	20 cm 8 in.
Adsorbent	Attapulgite
Adsorbent weight	15 kg 30 lb
Filter	
Area	214 cm^2 84 in.^{2}
Porosity	5 or 10 μm

DEGAS CIRCUIT (AND SURGE TANK):

Volume	95→135 l 25→35 gal(US)
Flow rate	12→63 ml/s 0.2→1.0 gal(US)/min
Heat removed	7 kW

The operating conditions, coolant conditions and the properties of the deposits are summarized in Table 2. Plots of the measured heat transfer coefficients are given in Figure 4.

A thin, black, very smooth deposit speckled with small discrete bumps was present on all heated surfaces. Figure 5 is a photograph of a portion of the surface of Section 1-2B. Micro-probe analyses of the deposits done by Euratom at Saluggia showed that the base film and the bumps had essentially the same composition. Tails of two distinct types were observed downstream of many of the bumps. One type was relatively short and dark in colour, the other was much longer and whiter in colour. Such bumps have not been observed in any subsequent tests.

3.1.2 Out-reactor Fouling Test No. 2

In preparation for Test No. 2, the loop was refilled with coolant from the dump tank. During circulation and filtering prior to the test, the pump seal failed catastrophically and approximately 60% of the coolant charge was lost. The charge was made up with more blended terphenyl-high-boiler mixture and filtered to bring the iron content down to less than 3 ppm.

Conditions for Test No. 2 were set to equal the surface temperatures and Reynolds numbers proposed for the in-reactor Test, X-706 (7). The bulk temperature was limited to 300°C, a lower value than proposed for the X-706 Test, to minimize the hazard of a pump seal failure until protective isolation valves could be installed. Part way through Test No. 2, hydrogen was added to the system to determine whether a steady hydrogen content could be maintained in the loop for proposed exposures of Zircaloy samples. During the test a continuous flow of approximately 0.1 l/sec was maintained through a 5 μm sintered stainless steel filter without plugging.

The coolant and operating conditions and the properties of the final deposit are summarized in Table 3.

The heated sections of the test sections were covered with a thin, smooth, black film, illustrated in Figure 6. The metallographic cross section of the film on section 2-3B shown in Figure 7, showed it to be a smooth, uniform apparently homogeneous layer, 11 μm thick well bonded to the test section surface. The film was mainly organic, the residue on ashing amounting to less than 10%.

TABLE 2

OUT-REACTOR TEST NO. 1

COOLANT AND OPERATING CONDITIONS, DEPOSIT OBSERVATIONS

COOLANT

Source: Santowax OM plus OMRE Core I HB, blended and rough filtered.

Time, hr	0	44	138 (Note 1)	139	215	354	476 (Note 2)	602	605
Iron content ppm	15	66	22	2	2	13	4	9	-
Water content ppm	-	-	-	-	-	-	-	176	-
PCFT, mg	28	-	30	61	-	-	-	-	316
Chlorine content ppm	5.2	2.1	2.3	2.4	-	-	-	-	0.5

Section	1-2A		1-2B		1-3A		1-3B	
OPERATING CONDITIONS								
Test duration (Note 2) hr.	476	149	476	149	476	149	476	149
Velocity, m/sec	1.5		1.5		3.7		3.7	
Reynolds no.	48,800		50,600		80,700		83,500	
Pressure at outlet (Note 2) g/cm^2	-	-	-	-	-	-	28	37
Bulk Temp. °C	344		351		357		364	
Surface temp, (Note 2) °C								
initial	478	504	505	536	471	489	499	527
final	481	500	508	536	467	488	498	525
Ht. tr. coeff, watts/cm^2 °C								
initial	0.230		0.232		0.487		0.450	
final	0.228		0.232		0.501		0.490	
Change in thermal resist., (watts/cm^2 °C)	0.04		0.00		-0.06		-0.18	
FINAL DEPOSIT (625 hr)								
Max. thickness, μm	<3		13		<3		8	
Weight, mg/cm^2	-		2.2		-		-	
Deposition rate, μg/cm^2 hr	-		4		-		-	
Residue on ignition, %	-		14		-		-	
Compounds by X-ray diff.	- - - - - - Fe_3O_4 and/or Fe_2O_3 - - - - - -							
Electrical resistance, arbitrary units	10^5		10^5		10^5		10^5	

NOTE 1: After 138 hr. of operation, power to test sections off for one week while filtering. Power instrumentation replaced.

NOTE 2: After 476 hr. of operation, power to test sections was increased. Valves given are for periods 0-476 hr and 476-625 hr.

FIGURE 4. FOULING TEST NO. 1

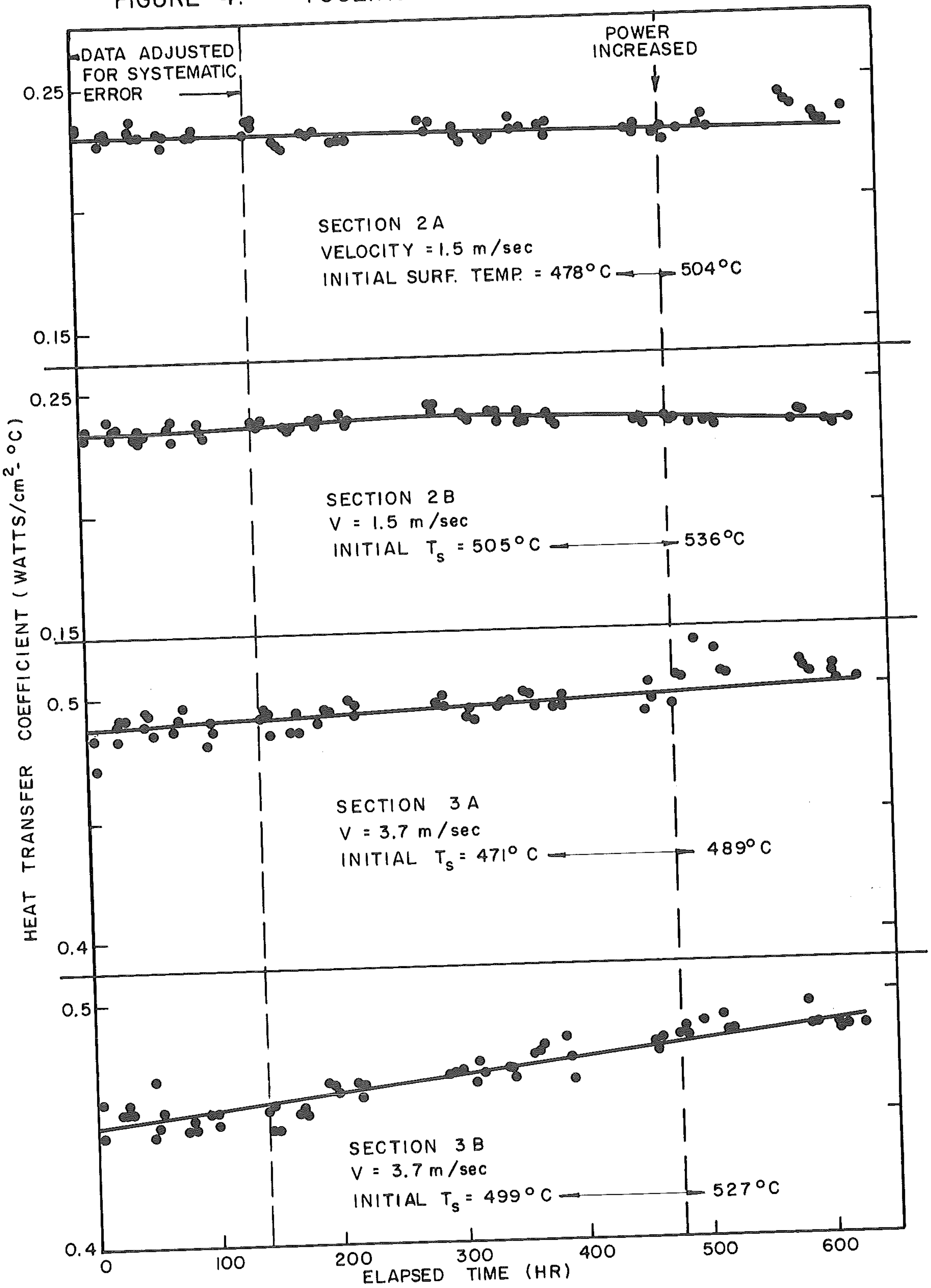


FIGURE 5. Deposit on Section 1-2B after 476 hr operation at T_s = 505°C plus 149 hr operation at T_s = 536°C and coolant velocity of 1.5 m/sec.

TABLE 3

OUT-REACTOR TEST NO. 2

COOLANT AND OPERATING CONDITIONS, DEPOSIT OBSERVATIONS

COOLANT

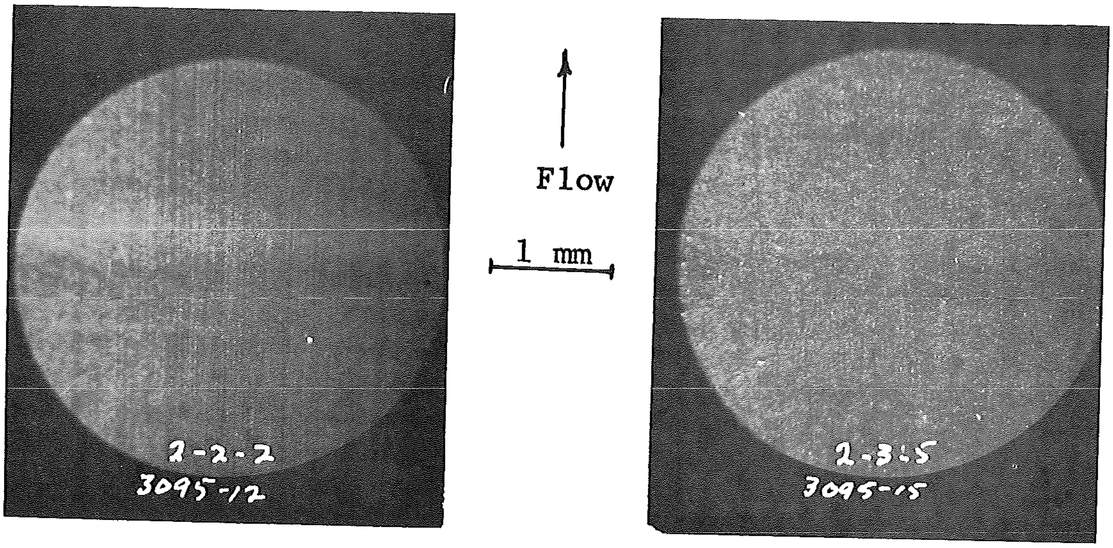
Source: 2 parts coolant from Test No. 1 combined with 3 parts blended OM-HB mixture, then recycled through 5 µm sintered stainless-steel filter.

Time, hr	0	169	193 (Note 1)	211	214	303	316	457	556
Iron content ppm	<3	<3	-	-	<3	<3	3	5	3
Water content ppm	215	-	-	-	-	-	-	-	213
PCFT, mg	50	-	56	-	-	-	99	-	45
Chlorine content ppm	2.0	-	-	-	-	-	-	-	1.3
Gas content, std cm^3/kg									
N_2	1289	-	-	949	-	963	-	-	734
H_2	0.3	-	-	22	-	39	-	-	26
CH_4	Trace	-	-	7.8	-	4.5	-	-	58
CO	-	-	-	2.5	-	nil	-	-	1.2
Hydrocarbons*	7	-	-	23	-	24	-	-	lost
Total	1296	-	-	1004	-	1031	-	-	-

Section	2-2A	2-2B	2-3A	2-3B
OPERATING CONDITIONS				
Test duration, hr	556	556	556	556
Velocity, m/sec	3.8	3.8	1.7	1.7
Reynolds no.	51,300	55,800	39,900	42,600
Pressure at outlet, kg/cm^2	-	-	-	27
Bulk Temp, °C	267	279	288	298
Surface Temp, °C				
initial	438	480	446	485
final	437	491	450	490
Ht. tr. coeff, $watts/cm^2$ °C				
initial	0.485	0.485	0.242	0.237
final	0.467	0.454	0.244	0.230
Change in thermal resist., $(watts/cm^2\ °C)^{-1}$	0.08	0.14	-0.03	0.13
FINAL DEPOSIT				
Max. thickness, µm	2	7	3	11
Weight, mg/cm^2	0.5	1.4	1.2	2.4
Deposition rate, $\mu g/cm^2$ hr	0.9	2.5	2.2	4.3
Residue on ignition, %	-	5	-	9
Compounds by X-ray diff.	- - - - - - - Fe_3O_4 and/or Fe_2O_3 - - - - - -			
Electrical resistance, arbitrary units	10^5	10^5	10^5	10^5

NOTE 1: Hydrogen added through sample bomb.

* Hydrocarbons = Compounds condensing between -77°C and -196°C at 0.005 mm Hg



Section 2-2B
T_s = 480°C
V = 3.8 m/sec

Section 2-3B
T_s = 485°C
V = 1.7 m/sec

FIGURE 6. Deposits in Out-reactor Test No. 2

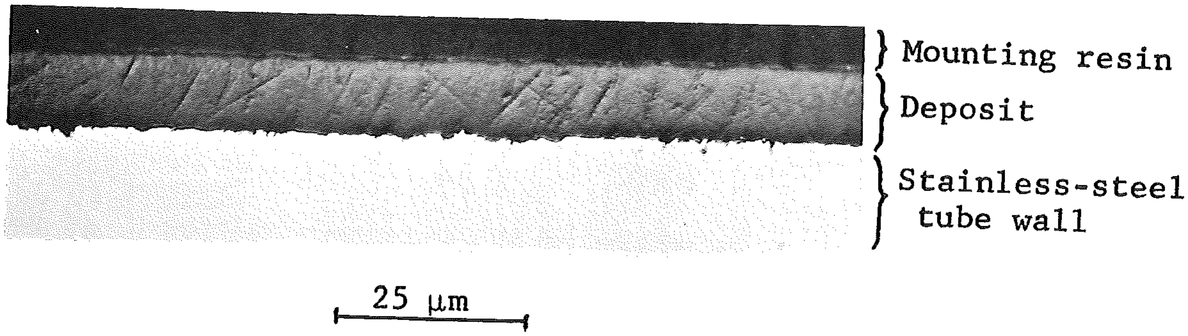


FIGURE 7. Cross Section through Deposit on Section 2-3B

The measured heat transfer coefficients showed little change during the run. The small changes observed on the two portions of section 2, occurred mainly during the first one-third of the run. The measured values are plotted in Figure No. 8.

3.1.3 Out-reactor Fouling Test No. 3

In preparation for Test No. 3, and in order to expose Zircaloy test specimens the coolant from Test No. 2 was circulated and filtered for one week at 370°C with a hydrogen content of 150 ml/kg. This coolant was drained from the loop while warm under an atmosphere of nitrogen. The loop was then filled with coolant removed from the X-7 loop, June 26, 1961, after completion of the X-706 Test (7). This coolant had been stored in a stainless steel drum at room temperature for 32 days. The coolant was circulated for a period of several days during initial calibration tests and Test No. 3 started. The coolant was not filtered, nor circulated through the surge tank at any time during the Test. In Test No. 3, conditions of surface temperature, bulk temperature and Reynolds number were made the same as existed in the X-706 test. The operating conditions, coolant conditions and the properties of the deposits are summarized in Table 4.

The heat transfer coefficients measured during the test are plotted in Figure 9. It will be seen that in Sections 2A and 2B with the lower velocity, the heat transfer coefficient decreased slowly. The heat transfer behaviour in Sections 3A and 3B was rather erratic. Both portions showed an initial sharp drop followed by a slower rise in heat transfer coefficient. Toward the end of the run the coefficients had apparently stabilized at approximately their initial values.

The marked difference in appearance of the deposits formed in the two test sections is evident in Figure 10. In Section 2 at the lower velocity the film was relatively smooth and had a matte finish. In the test section with the higher velocity (No. 3) the deposit was rather rough and sparkling. Close examination showed that the sparkle resulted from the crystal faces of many well-defined crystals imbedded in the film. These crystals showed equilateral triangle faces (Figure 11). The different structure of the two films was well illustrated in the metallographic cross-sections. (Figures 12 and 13). The deposits on the low velocity section were

FIGURE 8. FOULING TEST NO. 2

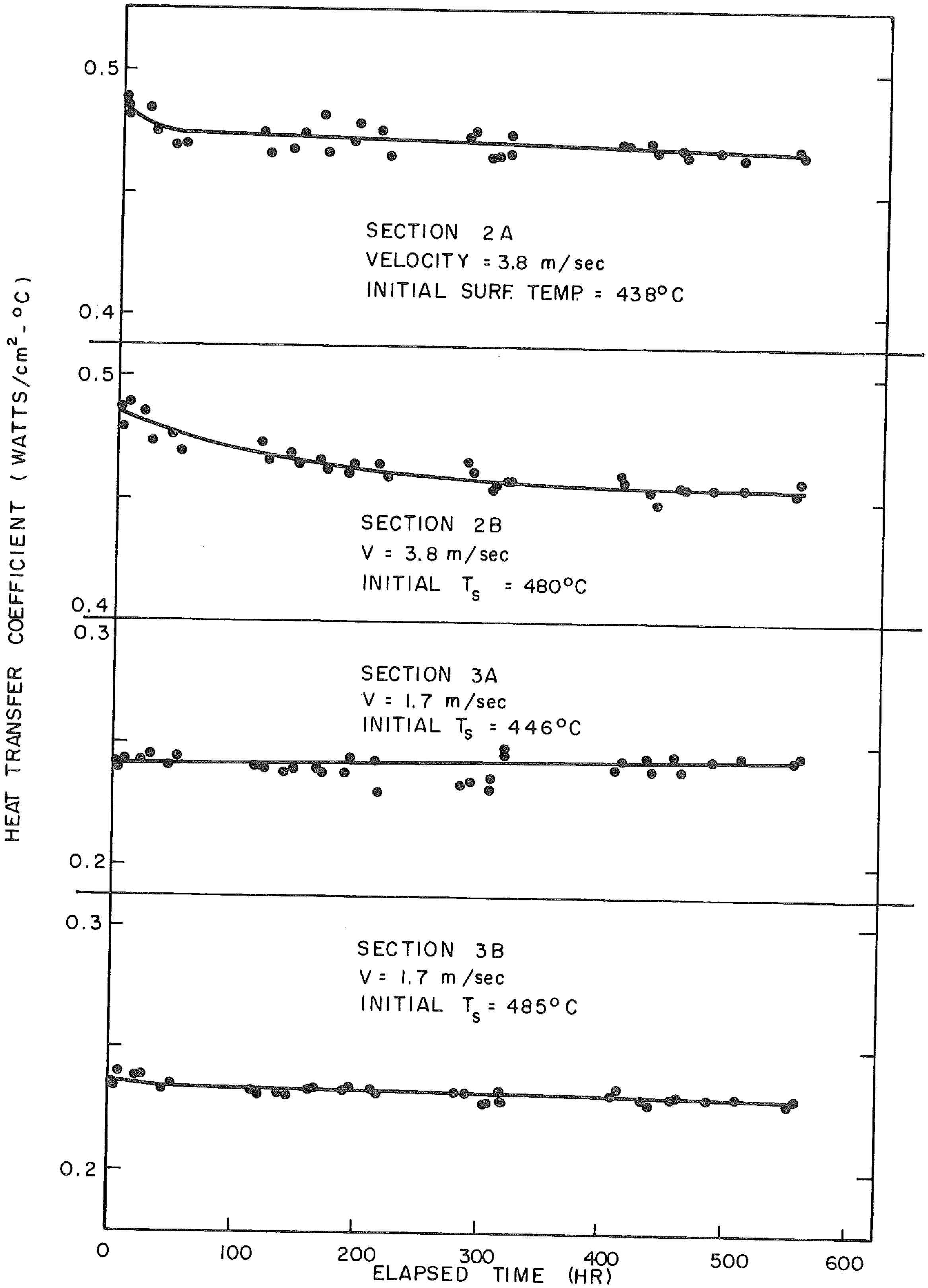


TABLE 4

COOLANT AND OPERATING CONDITIONS, DEPOSIT OBSERVATIONS

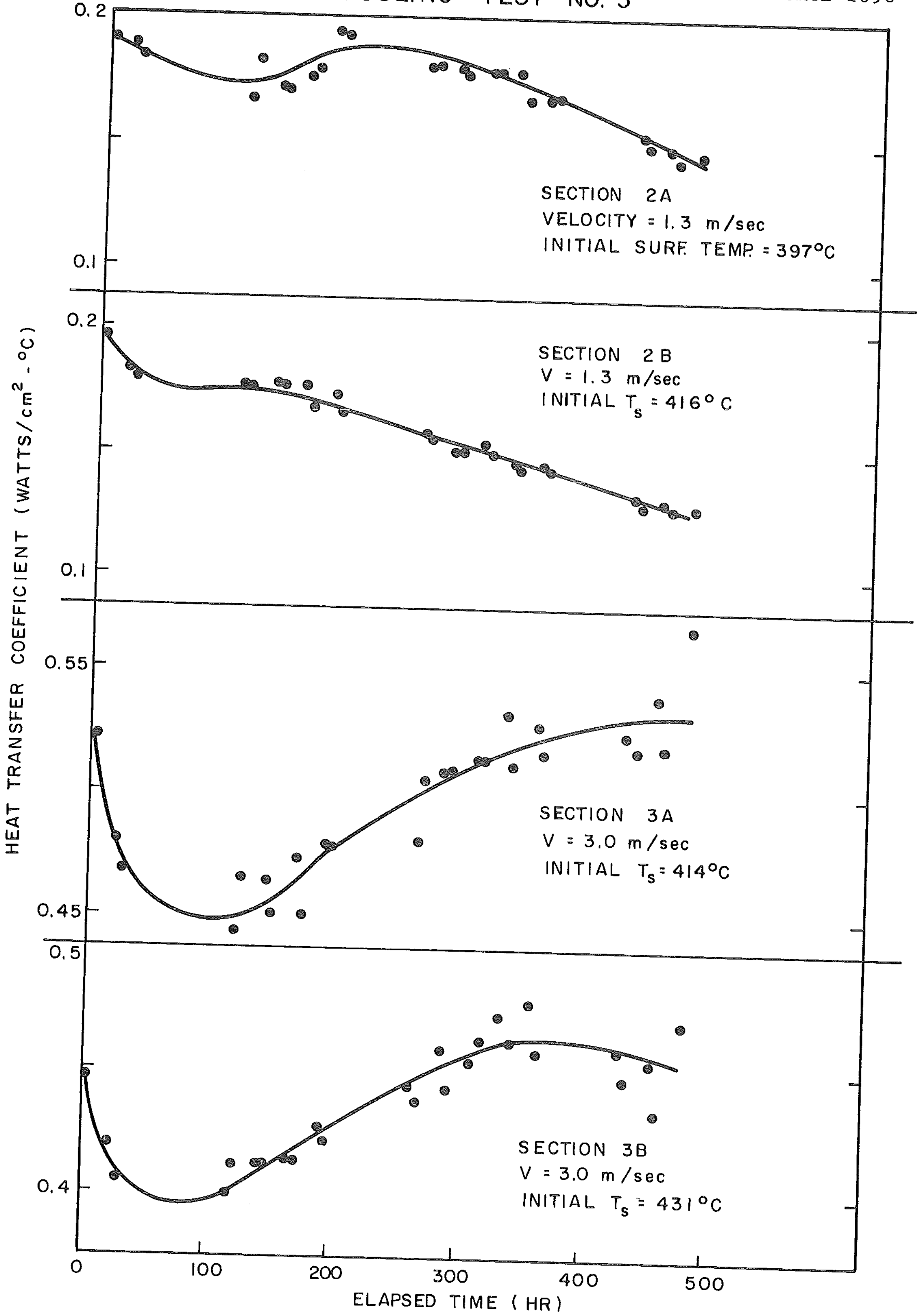
COOLANT

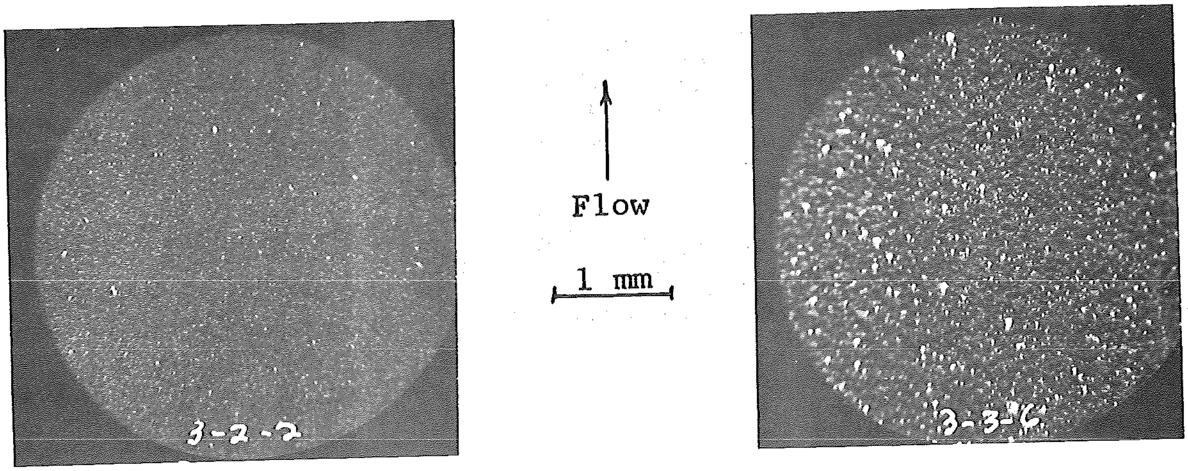
Source: Removed from X-7 Loop, 26/6/61

Time, hr	Before Test	0	192	195	314	459
Iron content ppm	6	3	-	2	2	2
Water content ppm	245	213	-	250	250	267
PCFT, mg	65	37	-	201	162	183
Chlorine content ppm	-	1.0	-	-	-	1.1
Gas content, std cm^3/kg						
N_2	-	-	281	-	-	-
H_2	-	-	50	-	-	-
CH_2	-	-	159	-	-	-
CO	-	-	1.3	-	-	-
Hydrocarbons	-	-	120	-	-	-
Total	-	-	611	-	-	-

Section	3-2A	3-2B	3-3A	3-3B
OPERATING CONDITIONS				
Test duration, hr	478	478	478	478
Velocity, m/sec	1.3	1.3	3.0	3.0
Reynolds no.	41,500	42,300	66,900	68,500
Pressure at outlet, kg/cm^2	-	-	-	14.8
Bulk Temp, °C	344	348	353	357
Surf. Temp, °C				
initial	397	416	414	431
final	434	451	404	426
Ht. tr. coeff, $watts/cm^2$ °C				
initial	0.193	0.197	0.523	0.450
final	0.143	0.127	0.533	0.455
Change in thermal resist., $(watts/cm^2\ °C)^{-1}$	1.8	2.8	-0.04	-0.02
FINAL DEPOSIT				
Max. thickness, µm	-	30	-	58
Weight, mg/cm^2	1.7	2.9	5.5	7.8
Deposition rate, $\mu g/cm^2$ hr	3.6	6.1	12	16
Residue on ignition, %	101	95	101	97
% Fe	-	55.0	-	67.0
% C	-	13.7	-	3.7
% H	-	0.7	-	0.3
Compounds by X-ray diff.	-	αFe, Pb, Fe_3O_4 and/or γFe_2O_3	-	Fe_3O_4 and/or γFe_2O_3, αFe
% Fe_3O_4 (calc'd)	-	55	-	96
% αFe (calc'd)	-	15	-	0
% Pb (calc'd)	-	16	-	0
Electrical resistance, arbitrary units				
inlet end	0.5	0.5	1	1
outlet end	1	1	1	1

FIGURE 9. FOULING TEST NO. 3





Section 3-2A
Ts = 397°C
v = 1.3 m/sec

Section 3-3B
Ts = 431°C
v = 3.0 m/sec

FIGURE 10. Deposits in Out-reactor Test No. 3

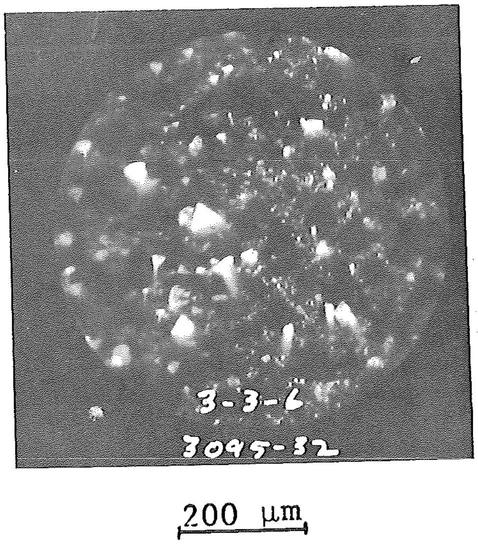


FIGURE 11. Higher Magnification View of Deposit on Section 3-3B showing well-defined Crystal.

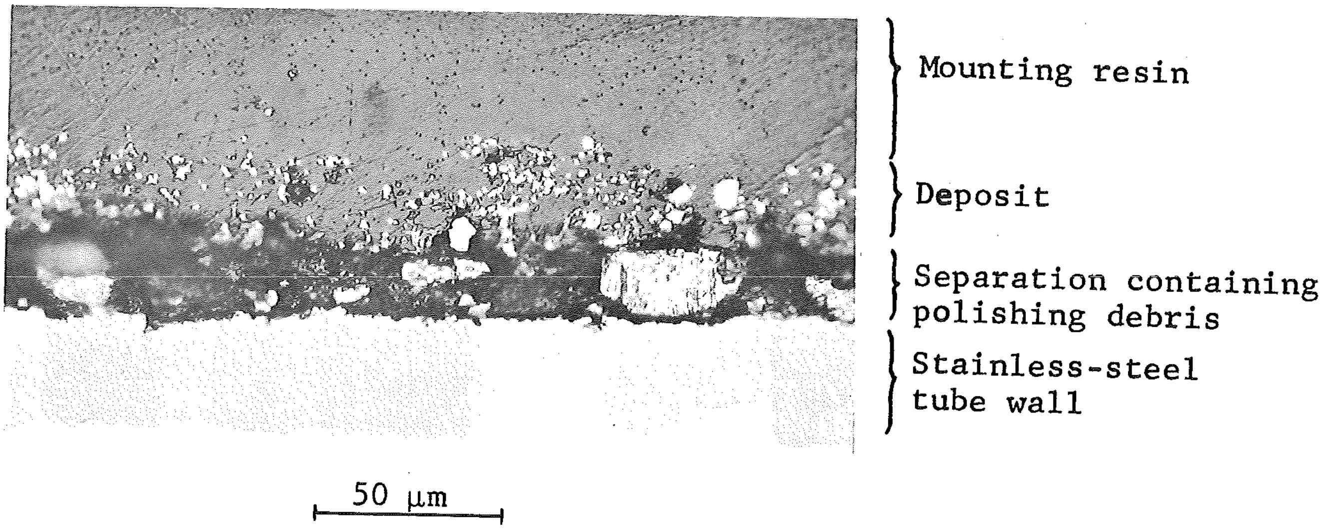


FIGURE 12. Cross Section of Deposit on Section 3-2B
T_s = 416°C; V = 1.3 m/sec

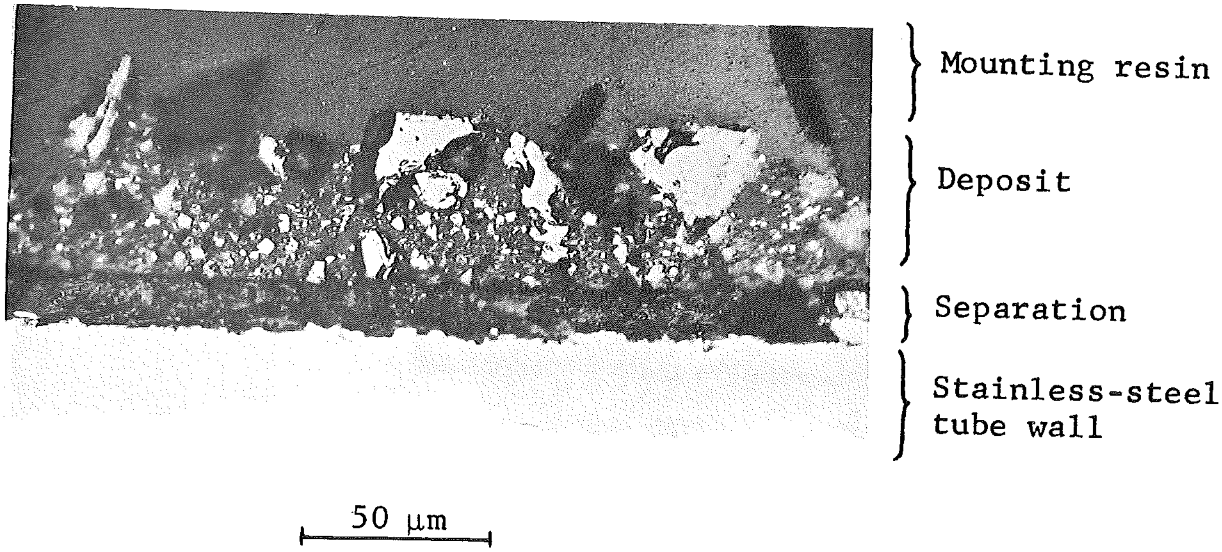


FIGURE 13. Cross Section of Deposit on Section 3-3B
T_s = 431°C; V = 3.0 m/sec

Note triangular crystal near right-hand side.

composed of fine particulates deposited in a rather porous layer. On the test section with the higher velocity the particles in the film were much larger and in some instances showed a regular triangular shape. In Figures 12 and 13, the dark band adjacent to the stainless steel is not part of the deposited film but rather a separation between the film and the base metal which occurred during the metallographic preparation. The large particles in this band are trapped polishing compound. The deposit on the test section with the higher velocity was both greater in amount and thicker than on the lower velocity test section. This was the first observed occurrence of an increased deposition rate with increased velocity.

Analysis of the deposits showed them to be predominantly inorganic. The inorganic portion was mainly magnetite plus some α-iron and elemental lead. The source of the lead was traced to pipe-joint compound used on two threaded joints in the loop. Its use was later discontinued. This test showed conclusively that the inorganic material in the film was not just that material present in the circulating coolant at the start of the test but rather was a result of a continuous mass transfer of material from the loop piping to the heater surfaces. The coolant in the loop contained a total of about 300 mg of inorganic material, measured as ash, at the start of the run and about 200 mg at the end. However, the deposits contained a total of 2200 mg. Another rather surprising observation was that the films were electrically conducting. Their electrical resistance, was orders of magnitude smaller than the films in Test No. 2.

3.1.4 Out-reactor Fouling Test No. 4

After Test No. 3 new test sections were installed and Test No. 4 followed immediately without draining the coolant from the loop. Conditions for Test No. 4, summarized in Table 5, were the same as in Test No. 3 except that the surface temperatures were raised about 16°C. This was designed to make the coolant pryolytic decomposition rate at the surface equal to the sum of pyrolytic and radiolytic rates at the surface of the fuel elements in the X-706 Test.

Changes in the heat transfer coefficient (Figure 14) measured for Test Section 4-2 in Test No. 4 were very similar to the changes observed for the corresponding section in Test No. 3. However, the values obtained in the higher velocity section did not follow the same pattern as was observed in the

TABLE 5

OUT-REACTOR TEST NO. 4

COOLANT AND OPERATING CONDITIONS, DEPOSIT OBSERVATIONS

COOLANT

Source: From Test No. 3

Time, hr	Before Test	0	314	388	478
Iron content ppm	2	<3	<3	-	3
Water content ppm	-	270	215	-	150
PCFT, mg	-	42	102	62	67
Chlorine content ppm	-	0.9	-	-	0.6
Gas content, std cm^3/kg					
N_2	-	-	-	-	175
H_2	-	-	-	-	55
CH_4	-	-	-	-	158
CO	-	-	-	-	0.9
Hydrocarbons	-	-	-	-	106
Total	-	-	-	-	494

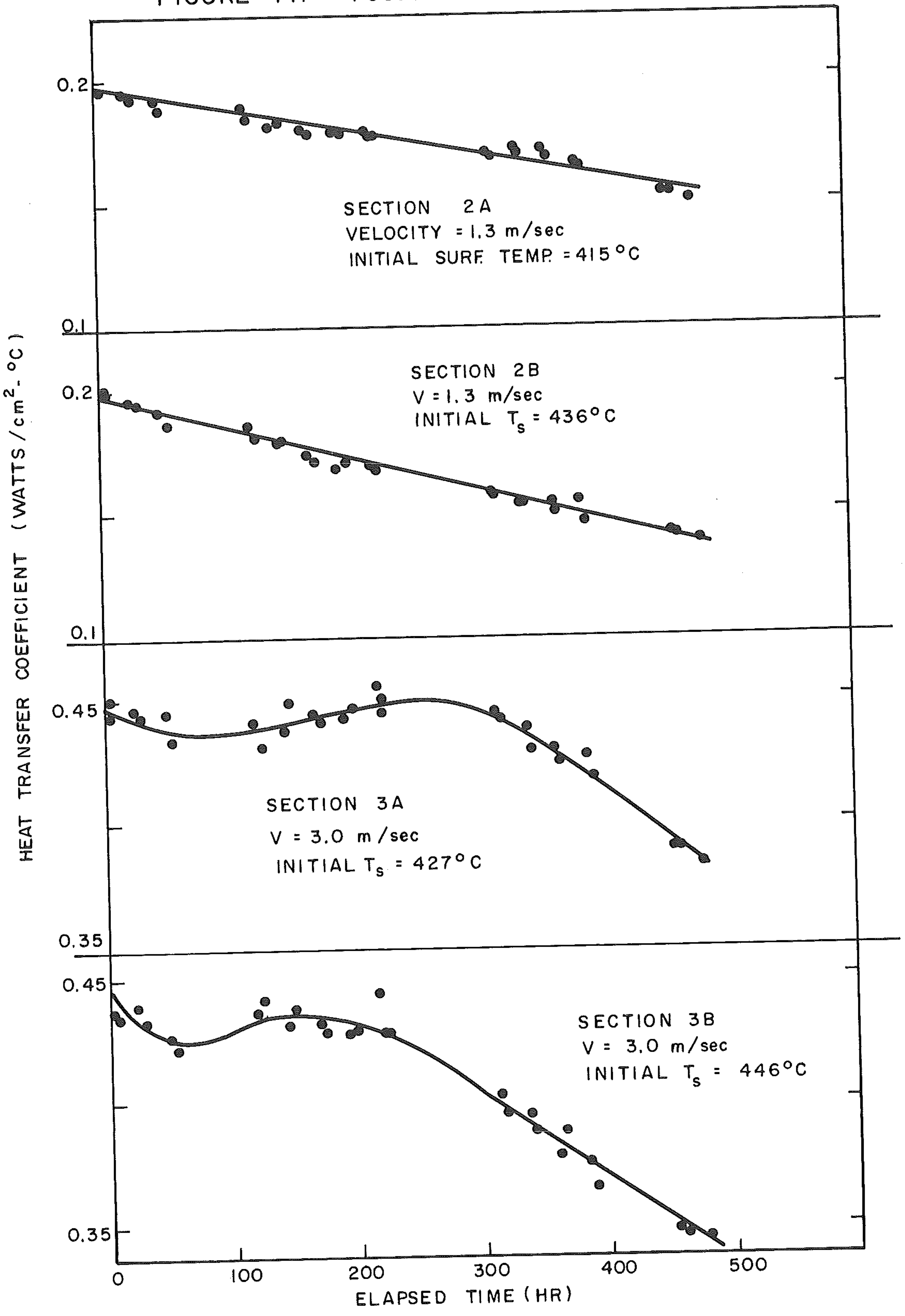
OPERATING CONDITIONS

Section	4-2A	4-2B	4-3A	4-3B
Test duration, hr	478	478	478	478
Velocity, m/sec	1.3	1.3	3.0	3.0
Reynolds no.	41,500	42,300	66,900	68,500
Pressure at outlet, kg/cm^2	-	-	-	14.4
Bulk Temp, °C	342	347	352	357
Surf. Temp, °C				
initial	415	436	427	446
final	446	483	433	463
Ht. tr. coeff, watts/cm^2 °C				
initial	0.198	0.197	0.448	0.445
final	0.154	0.138	0.383	0.343
Change in thermal resist., $(watts/cm^2\ °C)^{-1}$	1.4	2.2	0.4	0.7

FINAL DEPOSIT

Section	4-2A	4-2B	4-3A	4-3B
Max. thickness, μm	-	40	-	70
Weight, mg/cm^2	3.0	6.4	6.0	11.6
Deposition rate, $\mu g/cm^2$ hr	6.3	13	13	24
Residue on ignition %	88	60	104	96
% Fe	-	33.5	-	62.1
% C	-	44.9	-	17.1
% H	-	2.1	-	1.0
Compounds by X-ray diff.	-	Fe_3O_4 and/or γFe_2O_3, αFe, Pb	-	Fe_3O_4 and/or γFe_2O_3, αFe, Pb
% Fe_3O_4 (calc'd)	-	10	-	27
% αFe (calc'd)	-	32	-	48
% Pb (calc'd)	-	11	-	7
Electrical resistance, arbitrary units				
inlet end	0.5	3	0.5	1
outlet end	2	$>10^6$	1	3

FIGURE 14. FOULING TEST NO.4

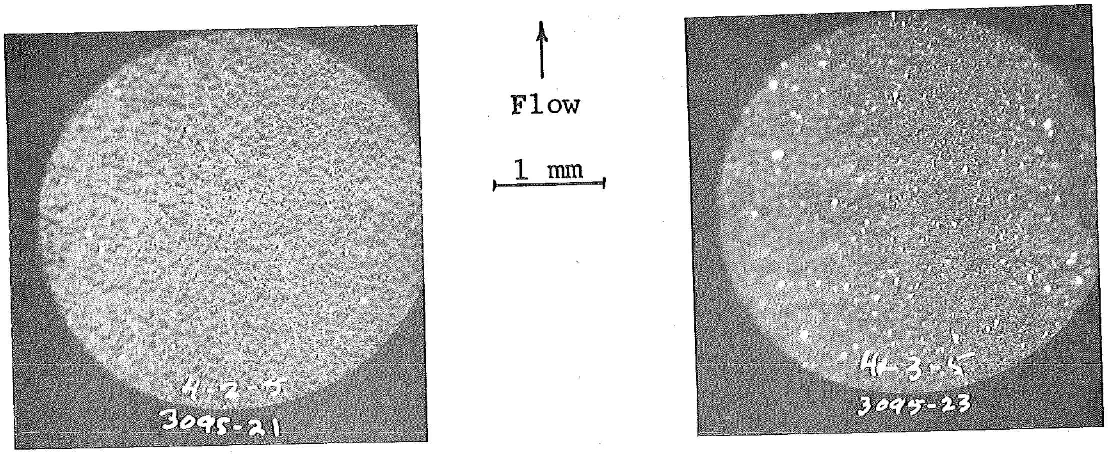


previous test. The heat transfer coefficients remained essentially constant for the first 250 hours after which they started a steady decline which persisted to the end of the run.

The properties of the deposited films are summarized in Table 5. Their appearance was similar to the corresponding films in Test No. 3, both microscopically as shown in Figure 15 and in cross section as shown in Figures 16 and 17. The deposits were, however, greater in weight and in thickness. Deposits in the lower velocity regions showed an appreciably higher weight loss on ignition indicating a greater organic content. The inorganic compounds present in the film were the same as in Test No. 3, i.e., Fe_3O_4 plus α-iron and lead. The total residue on ashing of the films was about 50% higher than in Test No. 3. The electrical resistance of the films was in general low, except in test section 4-2B. At the inlet end of this section the resistance was very low but increased sharply over the first cm to very high values. The film on the other test sections showed very slight increases in resistance toward the downstream end.

3.1.5 Out-reactor Test No. 5

Test No. 5 was a short-duration experiment to determine whether a complexing agent for iron, 8-hydroxyquinoline, when added to the coolant would prevent the deposition of iron compounds on heat transfer surfaces as had been indicated by early Phillips Petroleum work (8). Since only a yes-or-no answer was sought routine samples and heat transfer data were not taken. To the coolant remaining in the loop from Test No. 4 was added some additional make-up which had been removed from the X-7 loop June 26, 1961, after completion of the X-706 test. To this, 10 g (approximately 150 ppm) of 8-hydroxyquinoline was injected into the loop by means of the sample bomb and test No. 5 started. No filtering of the coolant either before or during the test was done. The test conditions are summarized in Table 6. During the period from 3 to 31 hours the bulk temperature gradually increased and the flow gradually decreased. This drift tripped the loop at 31 hours. The loop was then restarted and conditions held constant at initial values for the remainder of the run. At the time of the trip the bulk temperature was about 20°C high and the flow was about 10% low. Significant increases in the surface temperature of the test sections were observed and the test was terminated after 71 hours.



Section 4-2B
T_s = 436°C
V = 1.3m/sec

Section 4-3B
T_s = 446°C
V = 3.0m/sec

FIGURE 15. Deposits in Out-reactor Test No. 4

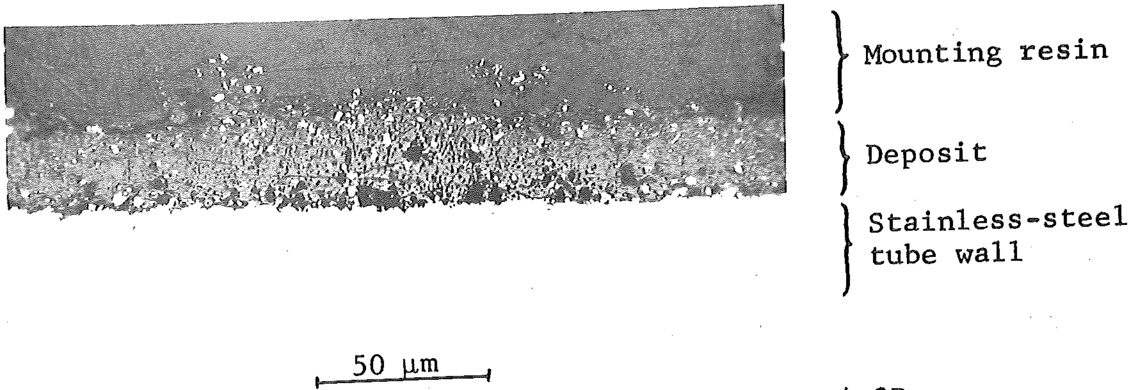


FIGURE 16. Cross Section of Deposit on Section 4-2B
T_s = 436°C; V = 1.3 m/sec

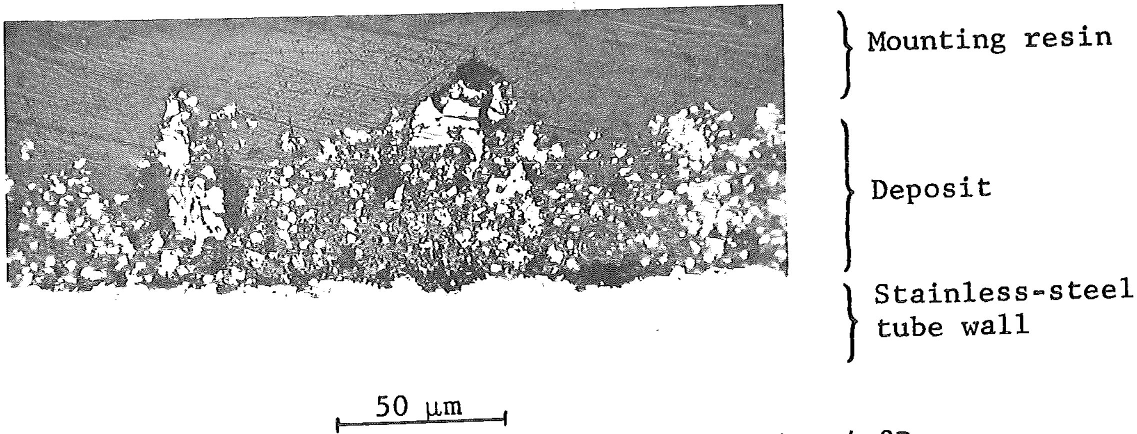


FIGURE 17. Cross Section of Deposit on Section 4-3B
T_s = 446°C; V = 3.0 m/sec

TABLE 6

OUT-REACTOR TEST NO. 5

OPERATING CONDITIONS, DEPOSIT OBSERVATIONS

COOLANT

Source: From Test No. 4 plus 10 g 8-hydroxyquinoline

Section	5-2A	5-2B	5-3A	5-3B
OPERATING CONDITIONS (Note 1)				
Test duration, hr	71	71	71	71
Velocity, m/sec	1.3	1.3	3.0	3.0
Reynolds no.	41,200	42,500	68,000	70,700
Pressure at outlet kg/cm^2	-	-	-	17.1
Bulk Temp, °C	343	349	356	364
Surface Temp, °C				
initial	452	484	453	482
final	-	523	-	531
Ht. tr. coeff, $watts/cm^2 °C$				
initial	-	0.202	-	0.457
final	-	0.158	-	0.323
Change in thermal resist., $(watts/cm^2\ °C)^{-1}$	-	1.4	-	0.9
FINAL DEPOSIT				
Thickness, μm	30	60	-	50
Weight, mg/cm^2	3.1	7.9	4.5	10.1
Deposition rate, $\mu g/cm^2$ hr	44	111	63	142
Residue on ignition, %	100	67	78	57
Compounds by X-ray diff.	-	αFe + Pb	-	αFe + Pb
Electrical resistance, arbitrary units				
inlet end	0	3	0	40
outlet end	1500	$>10^6$	1000	$>10^6$

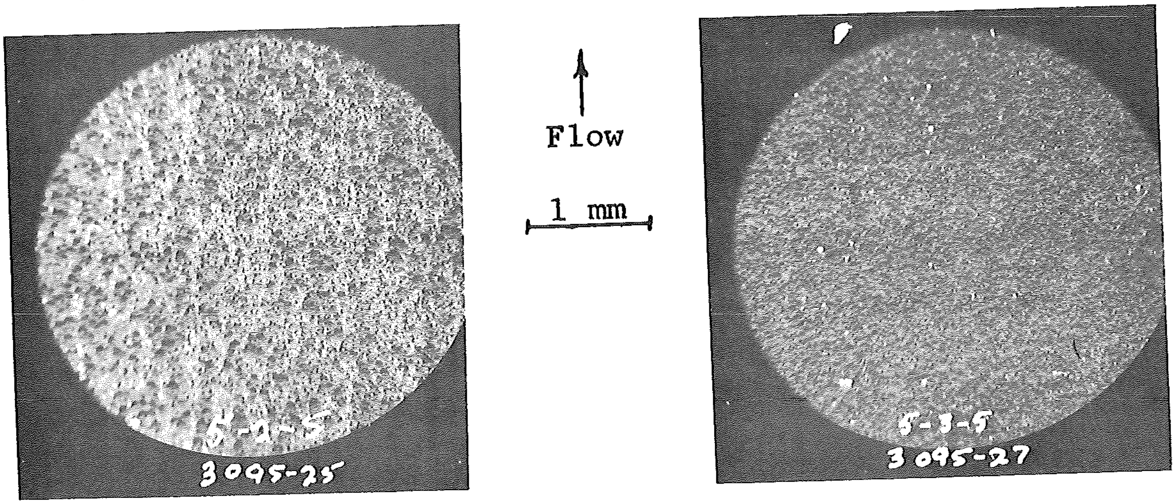
NOTE 1: Conditions drifted during period 3 to 31 hr. See text.

The heat transfer coefficients derived from recorder data and plotted in Figure 18 decreased steadily during the test. The coefficients plotted for the test have been normalized to the initial base conditions. Since there was no discontinuity in the coefficients across the loop trip at 31 hours the rapid drop immediately after restarting the test indicates a short period of very rapid deposition.

The only coolant analysis for Test No. 5 showed 14 ppm iron and 4.2 ppm chlorine in a sample taken after the loop had been cooled down at the end of the test.

The appearance of the films is shown in Figure 19. Analyses showed films from all sections except 5-2A to have an appreciable organic content. The inorganic portion consisted of α-iron and lead with no trace of oxides.

The electrical resistance of the deposits increased by several orders of magnitude from the inlet to the outlet end of each test section. On section 5-2A the resistance of the film rose exponentially from 0 to 1500 arbitrary units over the first half (15 cm) of the test section and was essentially constant over the remaining half. On section 5-2B it rose from 3 to greater than 10^6, the limit of the measurement, over the first 3 cm. Sections 5-3A and 5-3B were similar to 2A and 2B respectively except that on 5-3A the rise occurred over the full length of the section without any constant region.

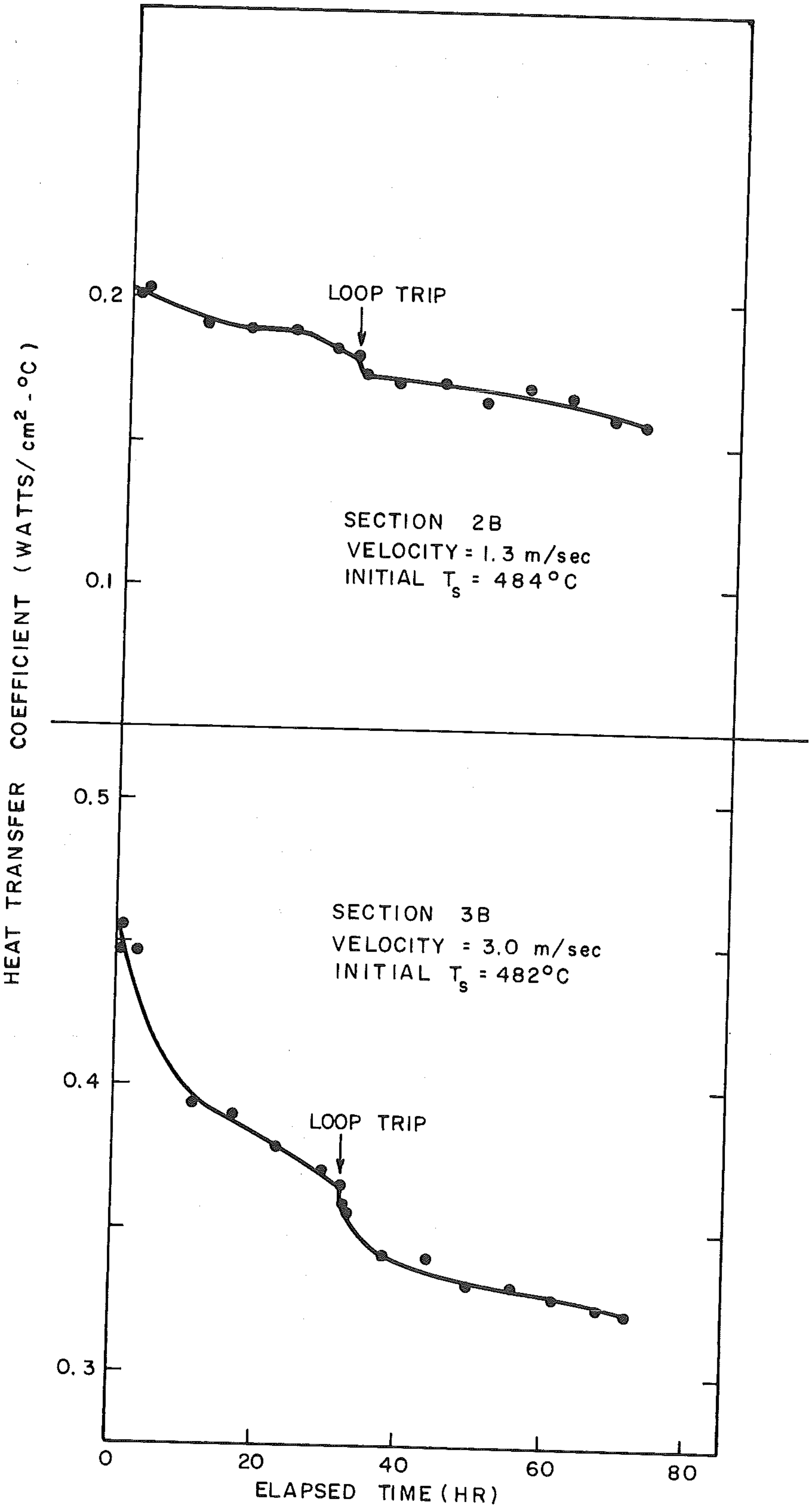


Section 5-2B
T_s = 484°C
V = 1.3 m/sec

Section 5-3B
T_s = 482°C
V = 3.0 m/sec

FIGURE 19. Deposits in Out-reactor Test No. 5

FIGURE 18. FOULING TEST NO. 5



This test showed no beneficial effects from the addition of the 8-hyroxyquinoline, as did a confirmatory second test by Phillips Petroleum (9).

3.1.6 Out-reactor Test No. 6

One of the most promising methods of coolant cleanup was thought to be treatment with Attapulgus clay. Test No. 6 was run to measure the fouling obtained with coolant which had been passed through an Attapulgus-clay bed to reduce its ash content and PCFT value (10) to relatively low values. Since Tests No. 3, 4 and 5 had shown greater deposits on the test section with the higher velocity, increased velocities were used in Test No. 6. The operating conditions are summarized in Table 7.

The coolant from Test No. 5 was drained from the loop and replaced by the coolant from Test No. 2; this coolant was circulated and filtered to flush the loop and drained. Coolant which had been purified by one pass through an Attapulgus clay column was charged to the loop and filtered. Although little filter plugging had occurred during the flushing operation seven filters were plugged with the new coolant before continuous operation could be obtained with the 5 μm filter and the test started. Filtration was continued throughout the test. The results of the coolant analyses are given in Table 7.

The measured heat transfer coefficients (Figure 20) decreased during the run in test sections 6-2A and 6-2B but increased sharply in test section 6-3A and 6-3B. These increases are believed to be mainly the result of the roughness of the deposit. The values were checked at the end of the run by reversing the individual power sources and power instrumentation between test sections 6-2 and 6-3. Raising the loop pressure temporarily from 20 to 27 kg/cm^2 caused no change in surface temperatures, thus confirming that boiling was not occurring.

The results of measurements and analyses of the deposited films are given in Table 7. As shown in Figure 21 the deposits on test sections 6-2A and 6-2B were quite smooth and fine grained, however, the deposits on test sections 6-3A and 6-3B were rough and coarse-grained. This difference is also evident in the metallographic cross sections in Figures 22, 23 and 24. The deposits especially on test section 6-2 were much thinner than in previous tests.

TABLE 7

OUT-REACTOR TEST NO. 6

COOLANT AND OPERATING CONDITIONS, DEPOSIT OBSERVATIONS

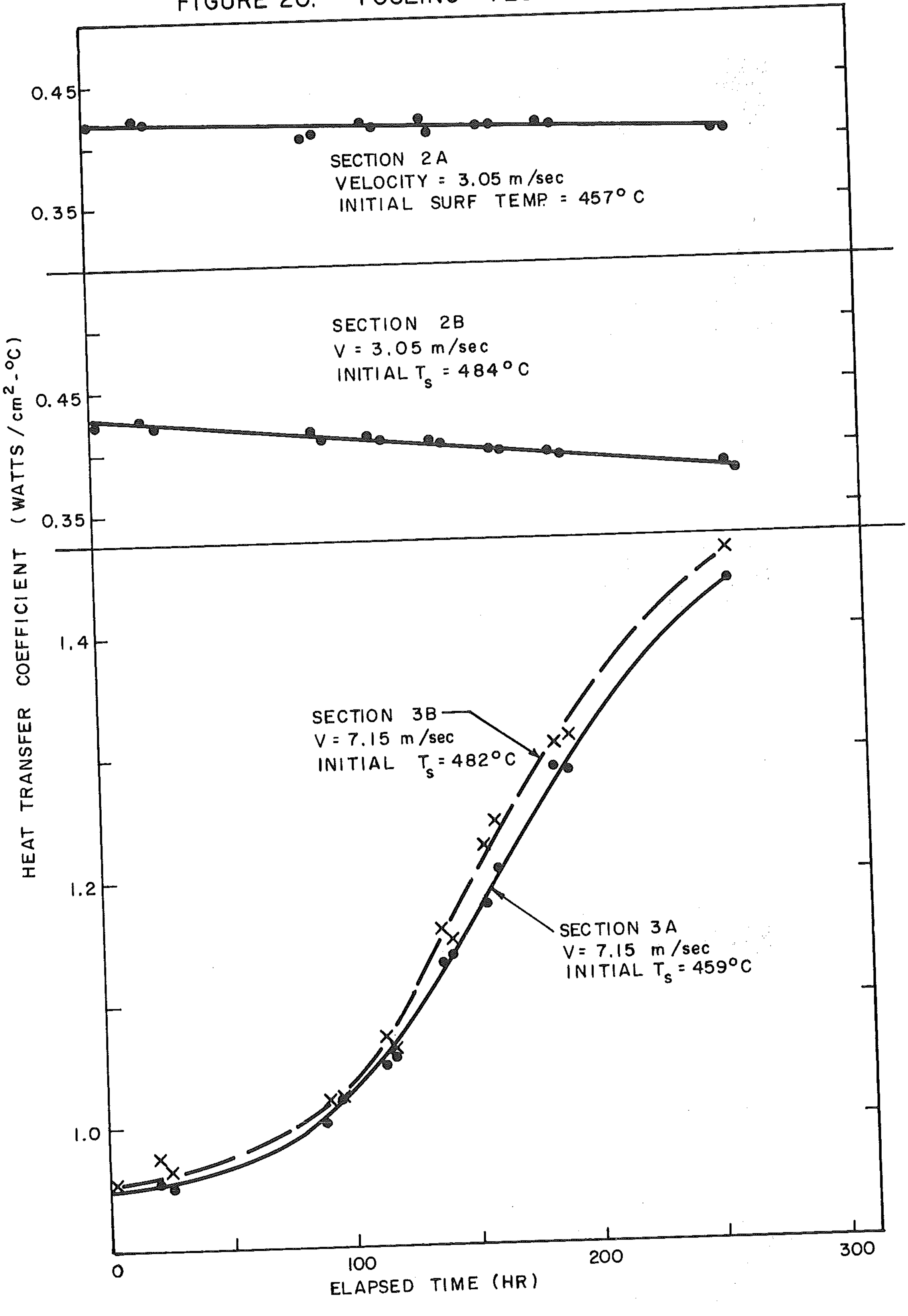
COOLANT

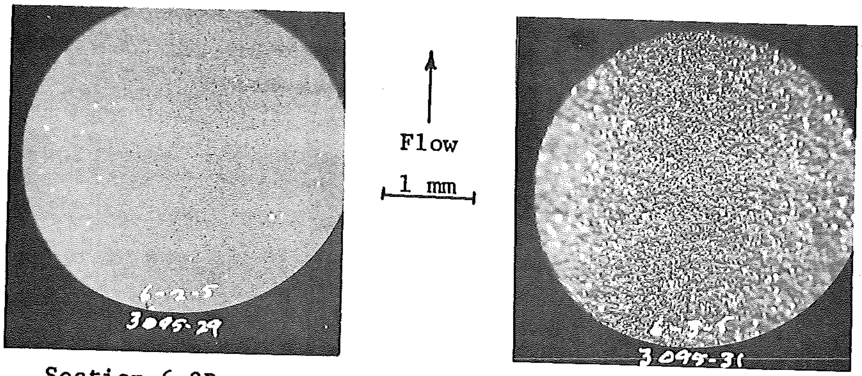
Source: Santowax OM plus OMRE HB purified by one pass through clay bed.

Time, hr	Before Charging	After Charging	-24	0	24	92	187	285
Iron content ppm	5	3	<2	<2	<2	<2	<2	<2
Water content ppm	-	-	90	-	-	-	-	84
PCFT, mg	8	121	8	7	7	9	18	18
Ash ppm	-	-	3	-	-	2	-	2
Chlorine content ppm	-	-	0.5	-	-	-	-	0.3
Gas content, std cm^3/kg								
N_2	-	-	-	243	-	-	-	462
H_2	-	-	-	57	-	-	-	74
CH_4	-	-	-	55	-	-	-	372
CO	-	-	-	0.1	-	-	-	0.1
Hydrocarbons	-	-	-	86	-	-	-	315
Total	-	-	-	441	-	-	-	1223

Section	6-2A	6-2B	6-3A	6-3B
OPERATING CONDITIONS				
Test duration, hr	281	281	281	281
Velocity, m/sec	3.1	3.1	7.2	7.2
Reynolds no.	98,000	101,000	161,000	166,000
Pressure at outlet, kg/cm^2	-	-	-	20
Bulk Temp, °C	345	352	358	365
Surface Temp, °C				
initial	457	484	459	482
final	460	501	423	442
Ht. tr. coeff, watts/cm^2 °C				
initial	0.421	0.429	0.952	0.955
final	0.406	0.379	1.44	1.47
Change in thermal resist., $(watts/cm^2\ °C)^{-1}$	0.09	0.30	-0.36	-0.37
FINAL DEPOSIT				
Max. thickness, μm	4	10	25	25
Weight, mg/cm^2	0.9	1.9	2.2	2.7
Deposition rate, $\mu g/cm^2$ hr	3.2	6.8	7.8	9.6
Residue on ignition %	-	98	109	101
% Fe	-	48.1	-	62.7
% C	-	11.7	-	2.4
% H	-	0.8	-	0.6
Compounds by X-ray diff.	-	αFe,Pb,Fe_3O_4 (trace)	-	Fe_3O_4, Pb (trace)
% Fe_3O_4 (calc'd)	-	41	-	90
% Fe (calc'd)	-	18	-	1
% Pb (calc'd)	-	29	-	7
Electrical resistance, arbitrary units				
inlet end	0	5	0	0
outlet end	1	100	0.5	0.5

FIGURE 20. FOULING TEST NO. 6





Section 6-2B
T_s = 484°C
V = 3.1 m/sec

Section 6-3B
T_s = 482°C
V = 7.2 m/sec

FIGURE 21. Deposits in Out-reactor Test No. 6

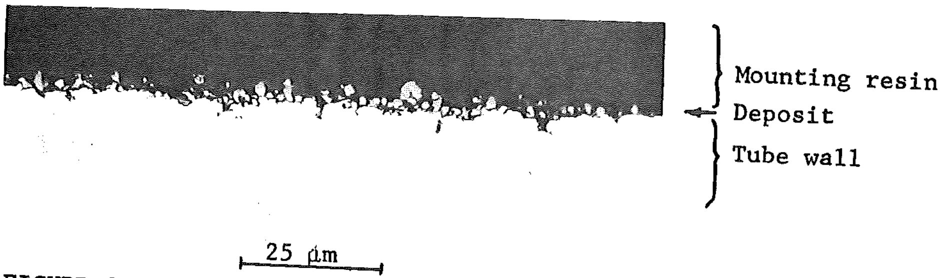


FIGURE 22. Cross Section of Deposit on Section 6-2A
T_s = 457°C; V = 3.1 m/sec

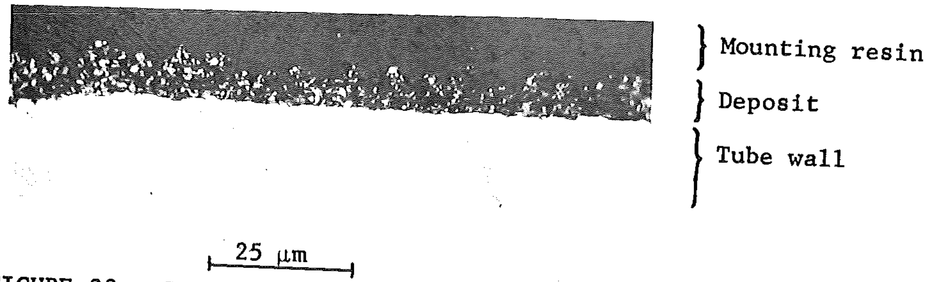


FIGURE 23. Cross Section of Deposit on Section 6-2B
T_s = 484°C; V = 3.1 m/sec

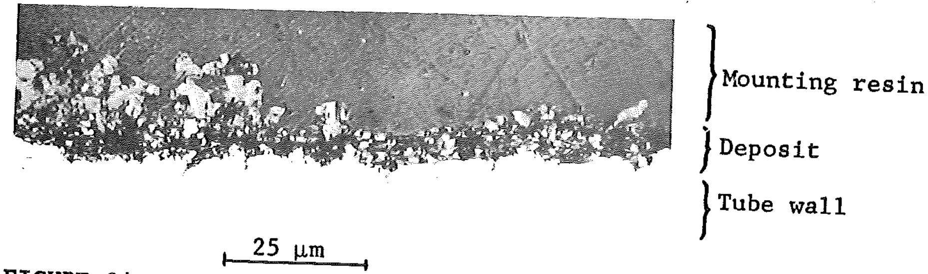


FIGURE 24. Cross Section of Deposit on Section 6-3B
T_s = 482°C; V = 7.2 m/sec
(Also representative of 6-2B, T_s=459°C; V=7.2 m/sec

X-ray diffraction analyses showed that the iron in the films was present as the oxide on test section 6-3B whereas it existed predominantly as alpha-iron on test section 6-2B. The organic content of the film was again small. The electrical resistance was also small but showed a small increase toward the outlet end of test section 6-2B.

3.1.7 Out-reactor Fouling Test No. 7

As a result of the encouraging results of Test No. 6 modifications were made to the loop to permit drying the coolant and continuously purifying it through an Attapulgus clay bed. Two 75 cm diameter by 90 cm long columns in parallel were installed upstream of the filter in the loop bypass. One column was charged with Linde Molecular Sieve LMS-4A and the other with pre-dried Attapulgus clay. The coolant from Test No. 6 plus some makeup was circulated through the LMS column at 100°C for about 24 hours to reduce its water content. The loop temperature was raised and after a period of filtering the coolant was passed through the clay column for about three days before starting the test. Both columns were operated with downflow at a rate of 70 l/hour (0.3 gal (US)/min) which is equivalent to a superficial mass velocity of 1500 g/hr cm^2, (3000 $lb/hr\text{-}ft^2$). Purification through the clay column was continued throughout the test. The operating conditions and coolant data are given in Table 8.

The heat transfer coefficients in test section 7-2 showed a barely significant decrease over the length of the run. Coefficients in test section 7-3 remained constant within the experimental error. The observed values are plotted in Figure 25.

The deposits were smooth films (Figure 26) too thin for thickness measurements. The only compound detected by X-ray diffraction was α-iron. The electrical resistance was too low to measure. The film weights compositions and deposition rates are also recorded in Table 8. High lead contents in the films again indicated contamination from the pipe-joint compound.

3.1.8 Out-reactor Fouling Test No. 8

In Fouling Test No. 7 which was run with a continuous purification of the coolant through the clay column and with a relatively low water content, the film deposition

TABLE 8

OUT-REACTOR TEST NO. 7

COOLANT AND OPERATING CONDITIONS, DEPOSIT OBSERVATIONS

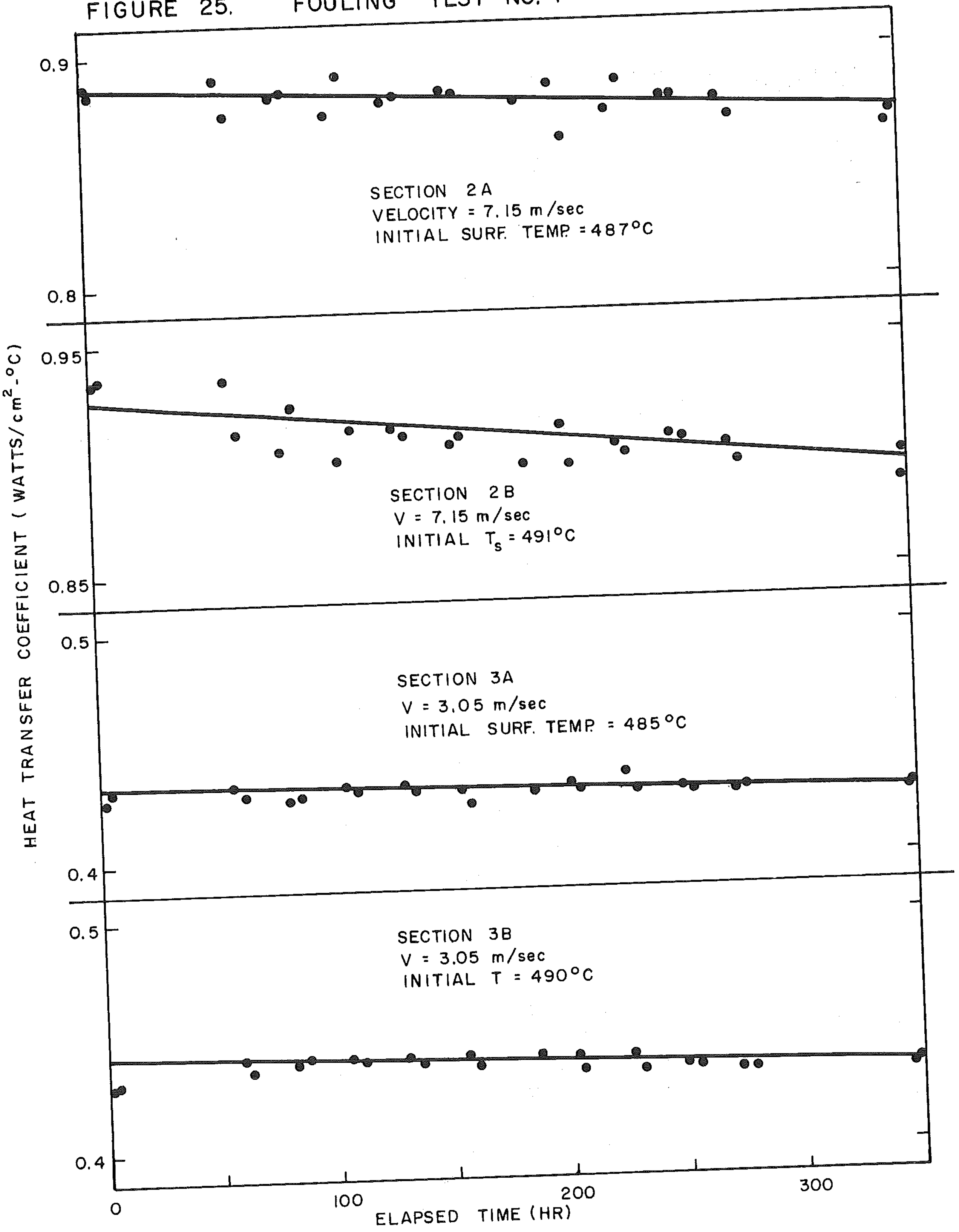
COOLANT

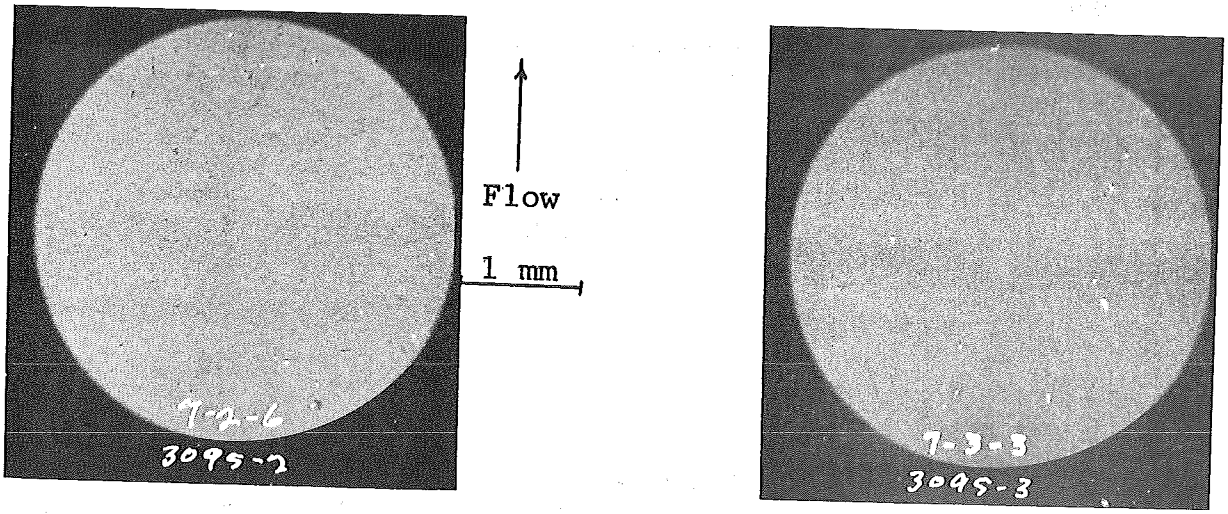
Source: From Test No. 6 plus clay-purified makeup.

Time, hr	After Charging	After Drying	-3	82	184	344
Iron content ppm	5	-	<2	-	-	-
Water content ppm	158	11	-	35	40	47
PCFT, mg	8	-	1	4	14	4
Ash content ppm	10	-	1	1	1	3
Chlorine content ppm	-	-	0.7	-	-	0.3
Gas content, std cm^3/kg						
N_2	-	-	294	-	-	-
H_2	-	-	48	-	-	-
CH_4	-	-	134	-	-	-
CO	-	-	1.4	-	-	-
Hydrocarbons	-	-	116	-	-	-
Total	-	-	593	-	-	-

Section	7-2A	7-2B	7-3A	7-3B
OPERATING CONDITIONS				
Test duration, hr	348	348	348	348
Velocity, m/sec	7.2	7.2	3.1	3.1
Reynolds no.	162,000	167,000	99,000	102,000
Pressure at outlet, kg/cm^2	-	-	-	17.7
Bulk Temp, °C	347	354	360	367
Surface Temp, °C				
initial	487	491	485	490
final	489	495	486	491
Ht. tr. coeff, $watts/cm^2$ °C				
initial	0.891	0.928	0.432	0.438
final	0.866	0.891	0.430	0.436
Change in thermal resist., $(watts/cm^2\ °C)^{-1}$	0.03	0.03	0.01	0.01
FINAL DEPOSIT				
Weight, mg/cm^2	0.3	0.3	0.2	0.3
Deposition rate, $\mu g/cm^2$ hr	0.9	0.9	0.6	0.9
Compounds by X-ray diff.	-	αFe + Pb	-	αFe + Pb
Electrical resistance, arbitrary units	0.5	0.5	0.5	0.5
Residue on ignition %	88	101	94	92
% Fe	30.2	34.1	14.9	17.7
% C	6.7	8.5	6.5	29.6
% H	3.1	3.1	1.1	3.9
% Pb	>25	>25	>25	>25

FIGURE 25. FOULING TEST NO. 7





Section 7-2B
Ts = 491°C
V = 7.2 m/sec

Section 7-3B
Ts = 490°C
V = 3.1 m/sec

FIGURE 26. Deposits in Out-reactor Test No. 7.

rate was about a factor of 15 less than in Test No. 6. In the latter test the coolant was clay treated before the run and only filtered during the test. Test No. 8 was conducted in order to determine whether the continuous purification or the low water content was principally responsible for the reduction in fouling rate. The test was conducted in three periods. The first period was run with continuous purification and a reduced water content; the second with continuous purification but a high water content; the third with no further purification and a high water content.

Before Test No. 8 was started, additional makeup coolant was added to the loop and the coolant re-dried with the molecular sieve column. The coolant was then circulated for about four days through the clay column which still contained the charge of clay used in Test No. 7. Three similar test sections had been installed in series in the loop. The reference test section having been replaced by a normal test section. With circulation continuing through the clay column at 100 l/hr, power was applied to test section 8-1. This portion of the test ran for 163 hours (not including a 2-1/2 hr interruption caused by a loop trip after 99 hours of operation). Makeup coolant and water equivalent to 300 ppm was added to the loop and power applied to test section 8-2. Flow through the clay column continued. After 25 hours of operation analytical results were received which showed a zero-time water content of only 100 ppm. Apparently the majority of the water was retained in the surge tank during initial mixing. Power to the test section was interrupted for a couple of hours while additional water was added to the loop. The test was resumed and then terminated after a total of 190 hours.

Since pump seal leakage was excessive throughout the test, another addition of makeup coolant from the dump tank was necessary before starting the third portion of the test. The flow through the adsorption column was continued for 3-1/2 hours to clean up the added coolant and then shut off for the remainder of the test. In order to remove excessive amounts of gas which had inadvertently been admitted from the dump tank during coolant addition, the loop was cooled to 150°C and circulation put through the surge tank. The loop temperature was then raised and additional water added. Power was applied to test section 8-3. During the subsequent 24 hours the coolant temperature gradually increased to 14°C above desired conditions and remained at this level an additional 24 hours at which time the loop tripped. The test was resumed at the original conditions and continued for a total of 140 hours. The operating conditions and coolant data for the three portions of the test are given in Table 9. The coolant analyses show a number of anomalies. They also show that some chlorine contamination was introduced at the start of the test. The reason for the increases in water content, PCFT, ash content and benzene-in soluble* content of the coolant during the first period is now known. Also, the iron and ash analyses are not consistent.

The measured heat transfer coefficients are plotted in Figure 27. In test section 8-1 the coefficients remain constant within experimental error. In test section 8-2, they remained constant for the first 140 hours and then increased for the remainder of the run. In test section 8-3 the coefficients increased steadily throughout the 140 hours of operation.

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* "Benzene insolubles" were determined on a series of coolant samples from Test No. 8 as part of an evaluation of the usefulness of the method for coolant characterization. "Benzene insolubles" denotes the percentage of the coolant which is retained by a 0.1 μm Millipore filter from a dilute benzene solution.

TABLE 9

OUT-REACTOR TEST NO. 8

COOLANT AND OPERATING CONDITIONS, DEPOSIT OBSERVATIONS

COOLANT	8-1				8-2						8-3			
Source:	From Test No. 7 + make-up, dried				From 8-1 + make-up + 35 g H_2O						From 8-2 + make-up + H_2O			
Time, hr.	-4 days	-5	96	161	-1	23 (note 1)	25	69	165	189	-5 (note 2)	-1	66	137
Iron content, ppm	8	<2	2	-	-	-	-	-	-	<2	2	7	2	-
Water content, ppm	18	60	63	53	102	116	314	212	214	-	160	286	-	190
PCFT, mg	31	8	44	60	20	-	-	7	5	4	-	11	-	9
Ash content, ppm	9	1	7	2	1	1	5	2	7	5	6	16	4	3
Benzene insolubles wt%	-	0.08	0.25	0.30	0.23*	0.27*	0.38*	0.31	0.47	0.45	0.47	0.41	0.67	0.72
Chlorine content, ppm	-	9.2	-	-	2.0	-	-	-	-	-	-	1.7	-	1.4

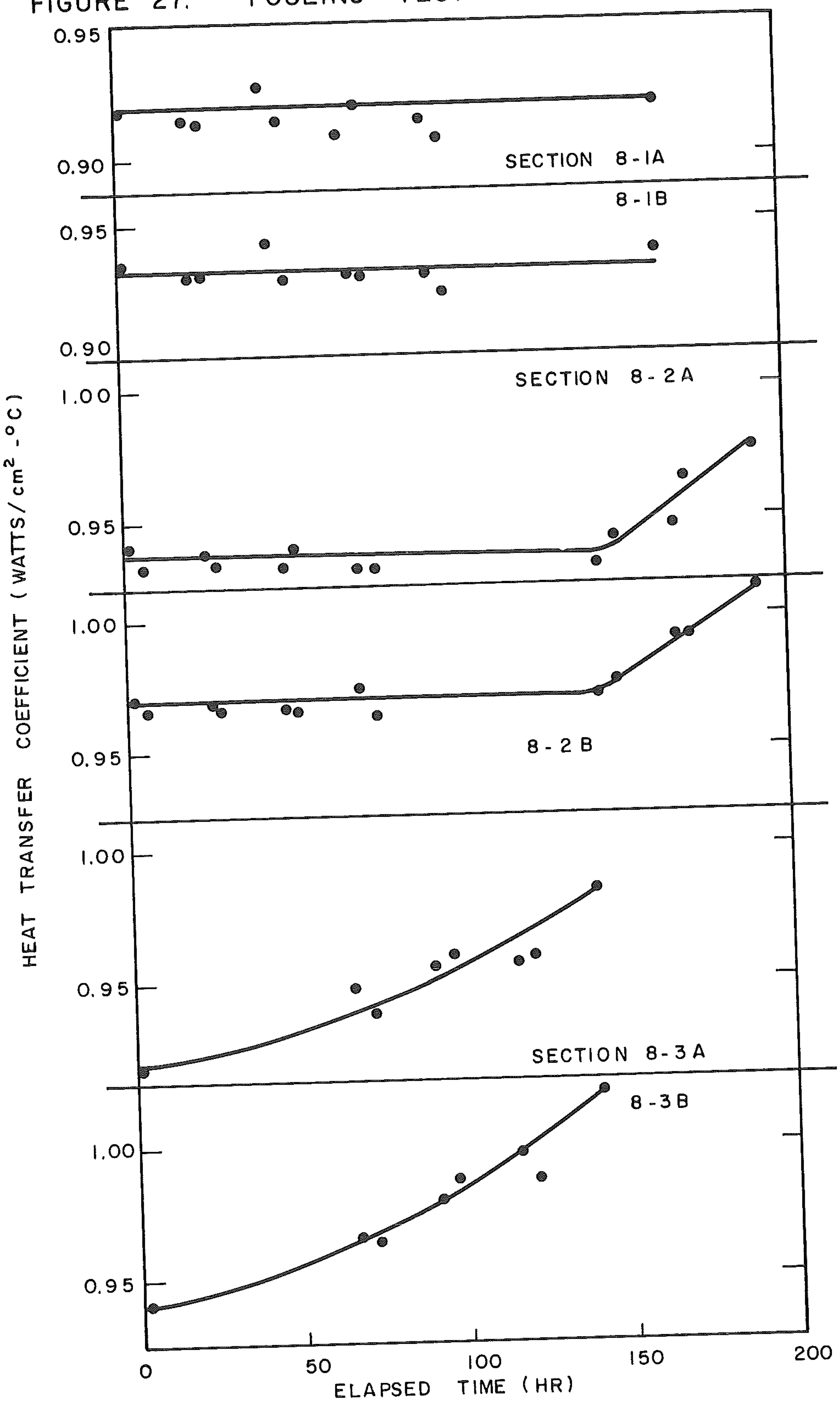
Section	8-1A	8-1B	8-2A	8-2B	8-3A	8-3B
OPERATING CONDITIONS						
Test duration, hr	163	163	190	190	140	140
Velocity, m/sec	7.2	7.2	7.2	7.2	7.2	7.2
Reynolds no.	162,000	167,000	162,000	169,000	162,000	169,000
Pressure at outlet kg/cm^2	-	18.2	-	18.2	-	16.9
Bulk Temp, °C	347	353	348	356	348	356
Surface Temp, °C						
initial	484	489	477	479	476	481
final	484	488	472	475	467	470
Ht. tr. coeff, watts/cm^2 °C						
initial	0.92	0.93	0.94	0.97	0.92	0.94
final	0.92	0.93	0.98	1.01	0.99	1.02
Change in thermal resist., (watts/cm^2 °C)$^{-1}$	0.0	0.0	-0.04	-0.04	-0.07	-0.08
FINAL DEPOSIT						
Weight, mg/cm^2	0.15	0.16	0.33	0.31	0.53	0.52
Deposition rate, μg/cm^2 hr	0.9	1.0	1.7	1.6	3.8	3.8
Compounds by X-ray diff.	-	-	Fe_3O_4+αFe	-	Fe_3O_4	-
Electrical resistance, arbitrary units	0.5	0.5	0.5	0.5	0.5	0.5
Residue on ignition %	-	98	-	95	99	-
% Fe	-	65	-	63	64	-
% C	-	2.1	-	3.1	5.1	-
% H	-	1.0	-	1.5	1.3	-

NOTE 1 24 g H_2O added after 23 hr.

NOTE 2 20 g H_2O added 4 hr before start of test.

* These results are of lower accuracy since the filter plugged when only 10-20% of solution had been filtered.

FIGURE 27. FOULING TEST NO. 8



The appearance of the deposits for the three periods of Test No. 8 can be compared from Figure 28. The deposit on test section 8-1 was very smooth, whereas the deposits on test section 8-2 and especially test section 8-3 were rougher and speckled with small bumps or agglomerates. These bumps are similar but smaller and less in number than those in Test No. 6 and are believed to be responsible for the increasing heat transfer coefficients. Why the increase did not start on test section 8-2 until after 140 hours of operation is not known. It is probable that the start of the increase is an indication of the chlorine becoming effective in promoting mass transfer, and masking, the effect of the changes in water content and purification. The film weight, deposition rate and results of the X-ray diffraction analyses are given in Table 9.

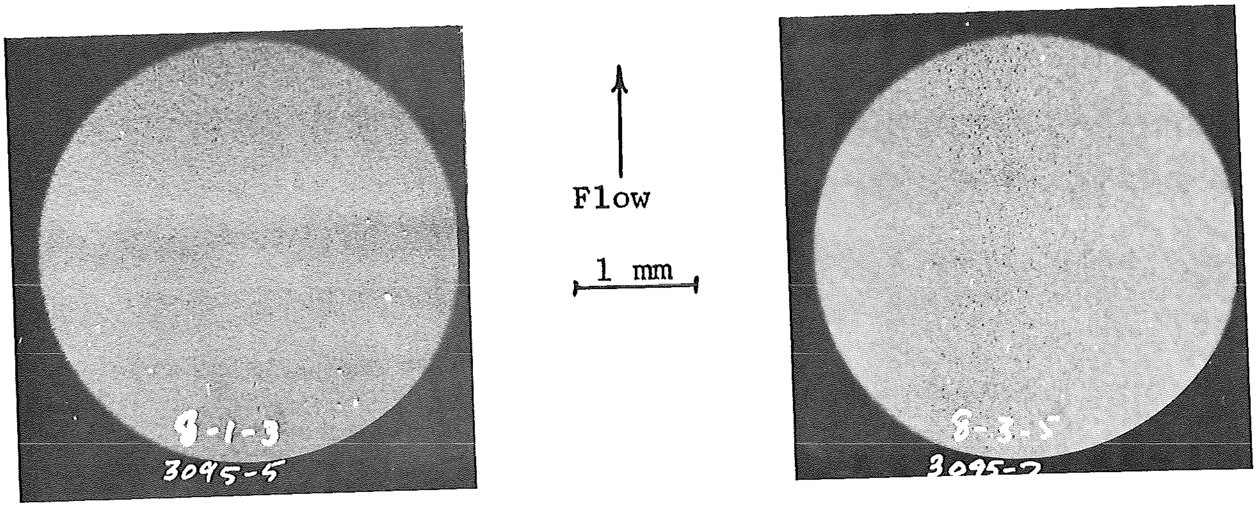
3.1.9 Out-reactor Test No. 9

Test No. 9 was planned as an extended run of at least six-weeks duration with well-cleaned coolant to determine fouling rates with a higher accuracy than was possible under similar conditions in the shorter tests No. 7 and 8-1. A range of surface temperatures (410°-495°C) was to be examined.

The starting coolant was a mixture of that remaining in the loop from Test No. 8 (40%) plus make-up coolant (60%), recently blended material which had been passed once through a partially spent clay column. In order to obtain well-purified and dried coolant a prolonged period (500 hours) of coolant treatment with LMS-4A Molecular Sieves and Attapulgus clay preceded the test. One new charge of LMS-4A, two new charges of clay and fifteen filter cartridges were required to bring the coolant to a condition which analyses indicated should result in a low fouling rate.

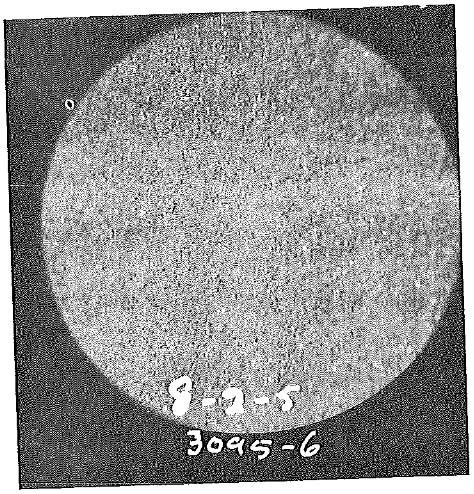
Less then one day after the test conditions given in Table 10 were established it was evident that fouling was occurring much more rapidly than would be predicted by the coolant analyses. The heat transfer coefficients remained within 6 per cent of the predicted values for about 7 hours and then increased. The measured values are plotted in Figure 29. The test was terminated after 44 hours.

Examination of the test sections showed the heat-transfer surfaces to be covered with a fine-grained deposit on which were superimposed sparkling bumps of coarser-grained material (Figure 30). The size and population of bumps



Section 8-1A
T_s = 484°C
V = 7.2 m/sec

Section 8-2B
T_s = 479°C
V = 7.2 m/sec



Section 8-3B
T_s = 481°C
V = 7.2 m/sec

FIGURE 28. Deposits in Out-reactor Test No. 8

TABLE 10

OUT-REACTOR TEST NO. 9

COOLANT AND OPERATING CONDITIONS, DEPOSIT OBSERVATIONS

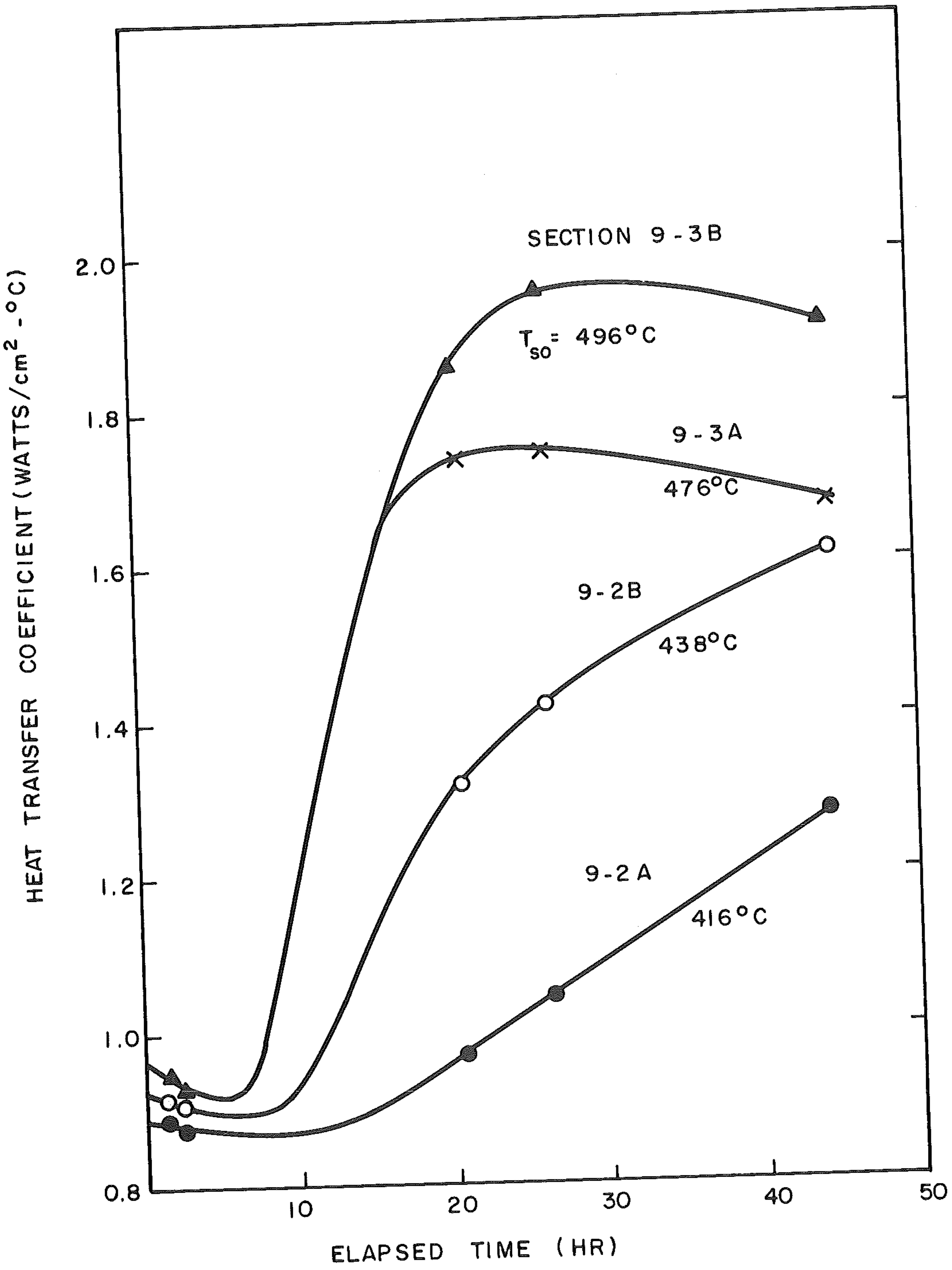
COOLANT

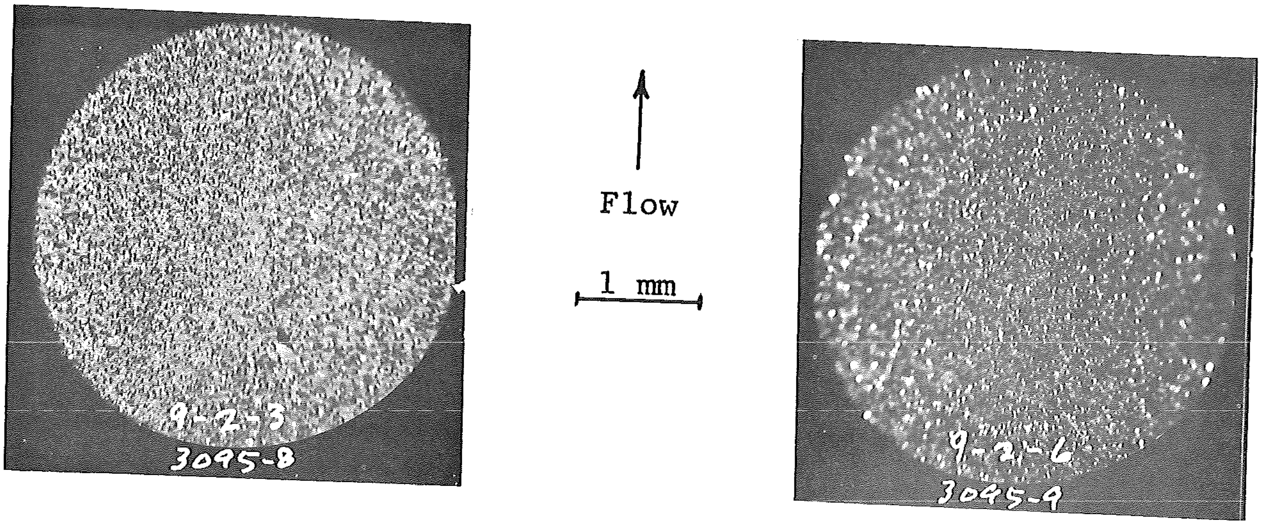
Source: 40% from Test 8-3, plus 60% partially cleaned makeup

Time, hr	-475	-425	-310	-220	-125	-52	-28	-4	44
Iron content ppm	-	20	-	-	-	1.5	-	3	2.5
Water content ppm	170	55	-	-	-	38	44	30	31
PCFT, mg	-	26	17	-	-	3	0	11	8
Ash content ppm	-	36	10	5	6	4	-	3	3
Benzene insolubles, wt %	-	-	-	-	-	0.17	-	0.23	0.38
Chlorine content, ppm	300	45	-	10	-	-	-	7.3	2.1
Gas content, std cm^3/kg									
N_2	-	-	-	-	-	-	-	193	-
H_2	-	-	-	-	-	-	-	73	-
CH_4	-	-	-	-	-	-	-	42	-
CO	-	-	-	-	-	-	-	0.9	-
Hydrocarbons	-	-	-	-	-	-	-	55	-
Total	-	-	-	-	-	-	-	364	-

Section	9-2A	9-2B	9-3A	9-3B
OPERATING CONDITIONS				
Test duration, hr	44	44	44	44
Velocity, m/sec	7.2	7.2	7.2	7.2
Reynolds no.	152,000	155,000	161,000	166,000
Pressure at outlet kg/cm^2	-	-	-	18
Bulk Temp, °C	345	351	357	365
Surface Temp, °C				
initial	416	438	476	496
final	393	400	423	429
Ht. tr. coeff, watts/cm^2 °C				
initial	0.89	0.92	0.94	0.95
final	1.28	1.61	1.67	1.90
Change in thermal resist., (watts/cm^2 °C)$^{-1}$	-0.34	-0.47	-0.46	-0.53
FINAL DEPOSIT				
Weight, mg/cm^2	0.79	2.12	4.87	6.42
Deposition rate, µg/cm^2 hr	18	47	109	144
Compounds by X-ray diff.	Fe_3O_4+αFe	Fe_3O_4+αFe	αFe+Fe_3O_4	αFe+Fe_3O_4
Electrical resistance, arbitrary units	<0.5	<0.5	<0.5	<0.5
Residue on ignition %				
% Fe	71.7	72.5	78.6	84.9
% C	-	0.4	0.6	1.0
% H	-	0.0	0.2	0.3
Metallic elements by emission spectroscopy	-	-	-	Cr - 0.02% Cu - 0.02 Ni - 0.07 Fe remainder

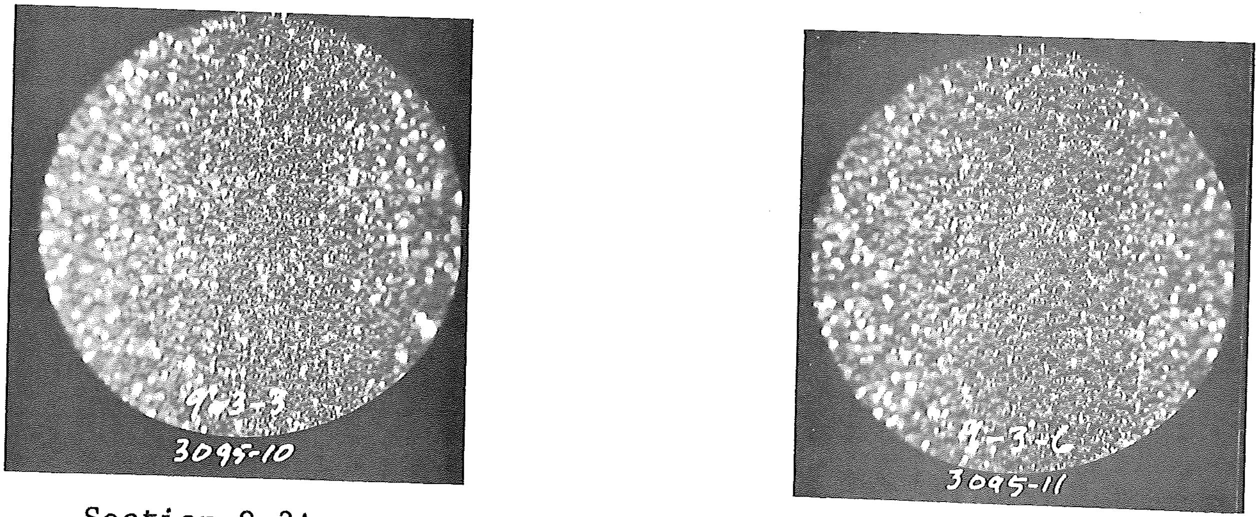
FIGURE 29. FOULING TEST NO. 9





Section 9-2A
T_s = 416°C
V = 7.2 m/sec

Section 9-2B
T_s = 438°C
V = 7.2 m/sec



Section 9-3A
T_s = 476°C
V = 7.2 m/sec

Section 9-3B
T_s = 496°C
V = 7.2 m/sec

FIGURE 30. Deposits in Out-reactor Test No. 9

increased with increasing initial surface temperature. Many triangular crystal faces could be observed in the coarse-grained material. The increases in heat-transfer coefficients are attributed to the roughness and additional surface area created by the deposits. The deposits contained negligible organic material. The results of measurements and analyses of the deposits are given in Table 10.

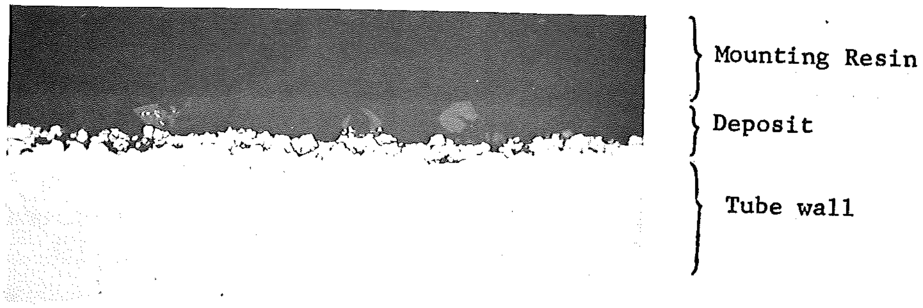
Metallographic cross sections (Figure 31) showed a sublayer of an approximately constant thickness of 8 μm on each section upon which was deposited a non-continuous layer of particles up to 30 μm across. The sublayer was identified as α-iron possibly with boundary layers containing Fe_3C and the large particles as iron oxide with αFe inclusions.

The deposition rates were a factor of about 150 times greater than would have been predicted from the coolant analyses and operating conditions based on the fouling rates in Tests No. 7 and 8.

Analyses received after completion of the test showed that the makeup coolant added before the test contained over 500 ppm chlorine, presumably from trichlorethylene contamination. The chlorine content, which on dilution in the loop was 300 ppm, had decreased to 45 ppm when the coolant temperature reached 340°C after treatment with the Molecular Sieve. It declined further to 10 ppm after 250 hours of circulation and 7 ppm at the start of the Test. The mechanism by which the chlorine was lost from the coolant is not known. It could either have reached with the mild steel or migrated to the cooler region in the surge tank. Later tests have shown that chlorine was responsible for the cleanup difficulties and the high fouling rates.

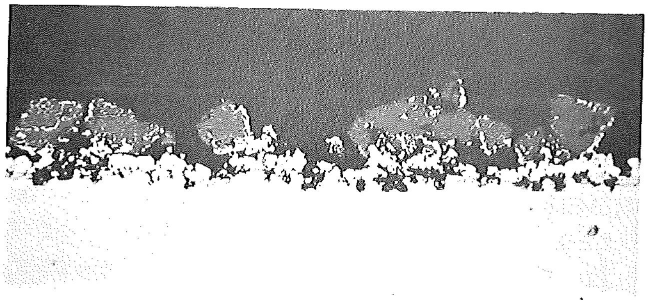
3.1.10 Summary of Out-reactor Tests

A summary of the data obtained in the nine out-reactor tests is given in Table 11.

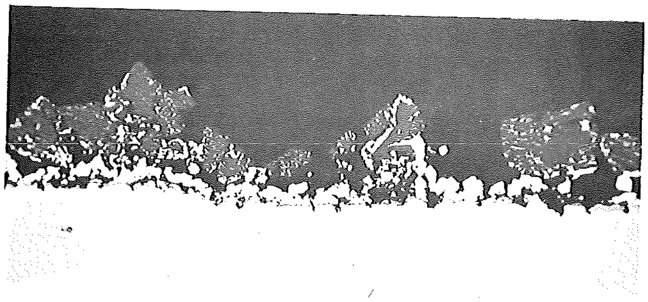


Section 9-2A T_s = 416°C

Section 9-2B T_s = 438°C



Section 9-3A T_s = 476°C



Section 9-3B T_s = 496°C

FIGURE 31 Cross Sections of Deposits in Out-reactor Test No. 9

50 µm V = 7.2 m/sec

TABLE 11

SUMMARY OF OUT-REACTOR FOULING TESTS

TEST NO.	1	2	3	4	5	6	7	8			9
COOLANT CONDITIONS											
Source	B	No. 1 + B	X-7	No. 3	No. 4 + X-7	B-SA	No. 6 + B-SA	No.7 + B-SA	No. 8-1	No. 8-2	No. 8 - 3 + B-SA
Pretreatment	F*	F	None	None	10g. 8-quinolinol added	F	D - RA - F	D - RA - F	W	W	D - RA - F
Treatment during Test	F*	F	None	None	None	F	RA - F	RA - F	RA - F	None	RA - F
Ash, ppm	-	-	-	-	-	2	1	3	4	6	3
Fe, ppm	16*	3	2	2	-	2	2	2	-	3	
Water, ppm	176	214	250	220	-	87	40	59	210	240	30
PCFT, mg	160*	65	150	70	-	13	6	37	9	10	9
Chlorine, ppm	2*	2	1	1	-	0.4	0.5	9*	2	1.5	5*

TEST SECTION	1-2A	1-2B	1-3A	1-3B	2-2A	2-2B	2-3A	2-3B	3-2A	3-2B	3-3A	3-3B
OPERATING CONDITIONS												
Test duration, hr	625	625	625	625	556	556	556	556	478	478	478	478
Velocity, m/sec	1.5	1.5	3.7	3.7	3.8	3.8	1.7	1.7	1.3	1.3	3.0	3.0
Initial Surf. Temp, °C	504*	536*	489*	527*	438	480	446	485	397	416	414	431
Coolant Temp, °C	344	351	357	364	267	279	288	298	344	348	353	357
DEPOSIT												
Texture	S	S+N	S	S+N	L	L	S	S	SX	SX	CX	CX
Max. thickness, μm	3	13	3	8	2	7	3	11	-	30	-	50
Deposition rate, $\mu g/cm^2$ hr	-	4	-	-	0.9	2.5	2.2	4.3	3.6	6.1	12	16
Change in thermal resist., $(watts/cm^2\ °C)^{-1}$	-	-	-	-	0.07	0.13	0.0	0.12	1.8	2.8	0.0	0.0
Residue on ignition, wt.%	-	14	-	-	-	5	-	9	101	92	101	100
Fe Compounds	-	0	-	-	-	0	-	0	0,I	0,I	0,I	0,I
Electrical resistance (arbitrary units)	10^6	10^6	10^6	10^6	10^6	10^6	10^6	10^6	1	3	1	1
Organic content, %(C+H)	-	-	-	-	-	-	-	-	-	14	-	4
Total Fe in deposit / Total Fe in coolant	~0.1				~0.3				11			

TEST SECTION	4-2A	4-2B	4-3A	4-3B	5-2A	5-2B	5-3A	5-3B	6-2A	6-2B	6-3A	6-3B
OPERATING CONDITIONS												
Test duration, hr	478	478	478	478	71	71	71	71	281	281	281	281
Velocity, m/sec	1.3	1.3	3.0	3.0	1.3	1.3	3.1	3.1	3.1	3.1	7.2	7.2
Initial Surf. Temp, °C	415	436	427	446	452	484	453	482	457	484	459	482
Coolant Temp, °C	342	347	352	357	343	349	356	364	345	352	358	365
DEPOSIT												
Texture	S	S	CX	CX	S	C	S	SX	S	S	CX	CX
Max. thickness, μm	-	40	-	70	30	60	-	50	4	10	25	25
Deposition rate, $\mu g/cm^2$ hr	6.3	13	13	24	44	111	63	142	3.2	6.8	7.8	9.6
Change in thermal resist., $(watts/cm^2\ °C)^{-1}$	1.8	2.2	0.4	0.6	-	1.4	-	0.9	0.09	0.30	-0.37	-0.38
Residue on ignition, wt.%	88	73	104	99	100	67	78	57	-	100	109	100
Fe Compounds	0,I	0,I	0,I	0,I	-	I	-	-	-	I,0	-	0
Electrical resistance (arbitrary units)	1	10^6	1	2	100*	>10^6	1000*	>10^6	0.5	40	0.5	0.5
Organic content, %(C+H)	-	47	-	18	-	-	-	-	-	13	-	3
Total Fe in deposit / Total Fe in coolant	13				-				6			

TEST SECTION	7-2A	7-2B	7-3A	7-3B	8-1A	8-1B	8-2A	8-2B	8-3A	8-3B	9-2A	9-2B	9-3A	9-3B
OPERATING CONDITIONS														
Test duration, hr	348	348	348	348	163	163	190	190	140	140	44	44	44	44
Velocity, m/sec	7.2	7.2	3.1	3.1	7.2	7.2	7.2	7.2	7.2	7.2	7.2	7.2	7.2	7.2
Initial Surf. Temp, °C	489	491	485	490	484	489	477	476	476	481	416	438	476	496
Coolant Temp, °C	347	354	360	367	347	353	348	356	348	356	345	351	357	365
DEPOSIT														
Texture	S	S	S	S	S	S	SX	SX	SX	SX	S+CX	CX	CX	CX
Max. thickness, μm	-	-	-	-	-	-	-	-	-	-	*	*	*	*
Deposition rate, $\mu g/cm^2$ hr	0.9	0.9	0.6	0.9	0.9	1.0	1.7	1.6	3.8	3.8	18	47	109	144
Change in thermal resist., $(watts/cm^2\ °C)^{-1}$	0.03	0.03	0.01	0.0	0.0	0.0	-0.04	-0.04	-0.07	-0.08	-0.34	-0.47	-0.46	-0.53
Residue on ignition, wt.%	-	-	-	-	-	-	-	-	-	-	-	106	111	119
Fe Compounds	-	I	-	I	-	-	-	0,I	-	0	0,I	0,I	0,I	0,I
Electrical resistance (arbitrary units)	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Organic content, %(C+H)	10	12	8	33	-	3	-	5	6	-	-	0.4	0.8	1.3
Total Fe in deposit / Total Fe in coolant	3				0.1		0.2		0.3		10			

SYMBOLS:

B	= Blended o-, m-terphenyl + 30% OMRE HB, rough filtered through 10 μm filter.
SA	= Single pass through Attapulgus clay bed.
F	= Recycled through 5 μm sintered stainless-steel filter.
RA	= Recycled through Attapulgus clay bed.
D	= Dried with Molecular Sieve bed.
W	= Water added.
X-7	= Removed from X-7 loop.
*	= Variable, see text for details.

DEPOSIT:

S	= Smooth, fine-grained.
C	= Coarse-grained.
L	= Very smooth, lacquer-like.
X	= Perceptible crystals.
N	= Nodules.
O	= Iron oxide.
I	= α-iron.

3.2 In-reactor Tests

Twelve fuel charges have been irradiated in the X-7 loop in NRX. The data obtained on fouling deposits on the fuel sheaths are in many cases much less quantitative and complete than in the out-reactor tests because of the difficulties in working with the highly radioactive specimens. The values summarized in Table 12 are representative averages of a range of values and are interpreted accordingly. Short descriptions of the tests and more details of the measurements are given in the following sections.

3.2.1 Commissioning Test X-57

Seven stainless steel sheathed UO_2 fuel elements mounted in a single string end-to-end were irradiated for two NRX cycles, a total of 44 days at power. This was the first fuel irradiation in the loop. Prior to this test, the loop had been operated out-reactor with terphenyl-high-boiler mixtures for a period of about one month. For about one week of this period the loop was operated at 425°C. Filtration through sintered stainless steel filters was used to clean up the coolant. At the start of the irradiation the coolant contained about 5 ppm iron. Iron contents throughout the run were in the range of 2 to 7 ppm.

All seven elements in the test showed fouling deposits over the fueled portions. The end caps were free of deposits. The boundary of the fouling at the downstream end of an element in a position corresponding to the end of the fuel is visible in Figure 32. The deposits exhibited two layers, an inner adherent hard layer and an outer powdery easily removable layer. The inner layer had 75% residue on ignition. The outer layer had 85%. X-ray diffraction analysis showed both layers to contain Fe_3O_4 and/or γFe_2O_3.

3.2.2 Zircaloy Feasibility Test X-67

This test was primarily to expose Zircaloy samples to organic coolant at 260°C. Two fuel elements similar to those used in the commissioning test were irradiated during the test. The deposits formed on the fuel sheaths were lacquer-like, much smoother and thinner than the films formed in the commissioning test. (See Figure 33). No analysis of the film was possible.

TABLE 12

SUMMARY OF X-7 IRRADIATIONS

TEST	FUEL ELEMENTS				AV.* IRRADIATION CONDITIONS				AV.* COOLANT CONDITIONS			DEPOSIT*		
	Arrangement	No. of Elements	Sheath	Fuel	Duration Days	Velocity m/sec	Bulk Temp., °C	Surface Temp., °C	Iron ppm	Water ppm	PCFT mg	Wt/unit area mg/cm^2	Thickness µm	% Residue on ignition
X-57 Commissioning	Single string	7	S.S.	UO_2	44	9	385	460	3	-	-	-	25-100	80
X-67 Zircaloy Feasibility	Single string	2	S.S.	UO_2	20	9	260	370	3	-	-	-	<10	-
X-58 First SAP	Single string	7	SAP	UO_2	0.3	9	385	475	18	-	-	-	<5	-
X-59 SAP Defect	Single	1	SAP	UO_2	15	9	370	475	3	-	-	-	<10	-
X-708 Finned Small Element	Single string	7	SAP	UO_2	30	9-4.5	370	435	3	230	75	-	20-35	-
X-704-1 First Trefoil, Phase 1	2 Trefoils	6	SAP	UO_2 }	1	8	330	420	7	190	-	-	10-500	34-90
	1 Trefoil	3	S.S.	UO_2 }										
X-704-2, First Trefoil, Phase 2	1 Trefoil	3	SAP	UO_2 }	32	8	340	430	4	150	70	-	10-1400	53
	1 Trefoil	3	S.S.	UO_2 }										
X-706, Fuel Element Fouling	Single String	4	S.S.	Pu-Al	21	2.5	347	400	3	170	90	7	0-40	90
		3	S.S.	Pu-Al	21	7	353	430	3	170	90	17	0-50	90
X-709-1, Long Term Trefoil, Phase 1	2 Trefoils	6	SAP	UO_2	76	7	310	450	3	170	10	-	40-80	70
X-707 Beryllium-Zircaloy Clad	Single string	10	8-Be; 2 Zr.	UO_2	20	9	375	450	4	50	6	0.2	<5	-
X-711 Uranium Carbide	Single string	8	6 SAP; 2 S.S.	UC	8	8	340	410	2	30	4	-	<5	-
X-709-2 Long term Trefoil, Phase 2	2 Trefoils	6	SAP	UO_2	0.8	7	295	420	2	35	4	0.02	<5	-

* NOTE: The values given in this table are, in many cases, representative averages.

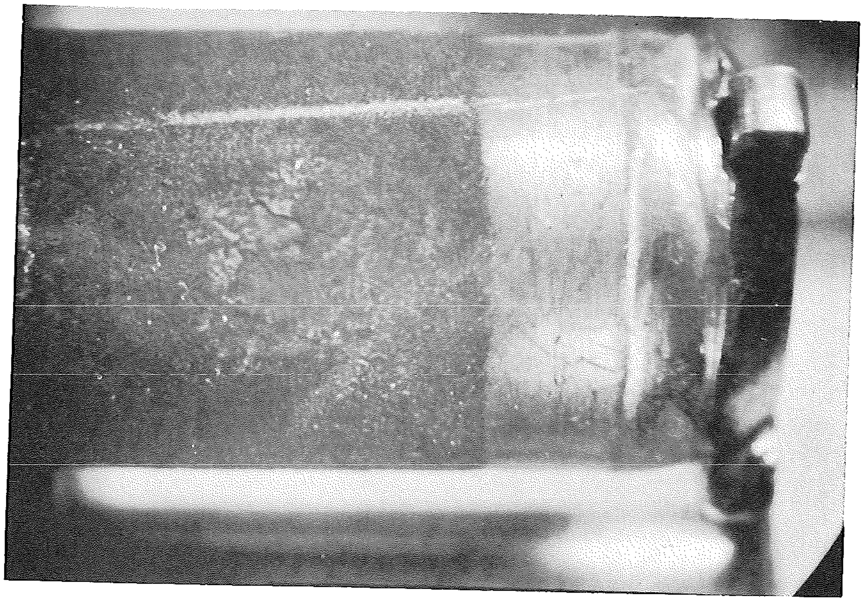


FIGURE 32. Downstream end of fuel element from Commissioning Test, X-57.
$T_s \sim 450°C$ $V = 9$ m/sec

1 cm

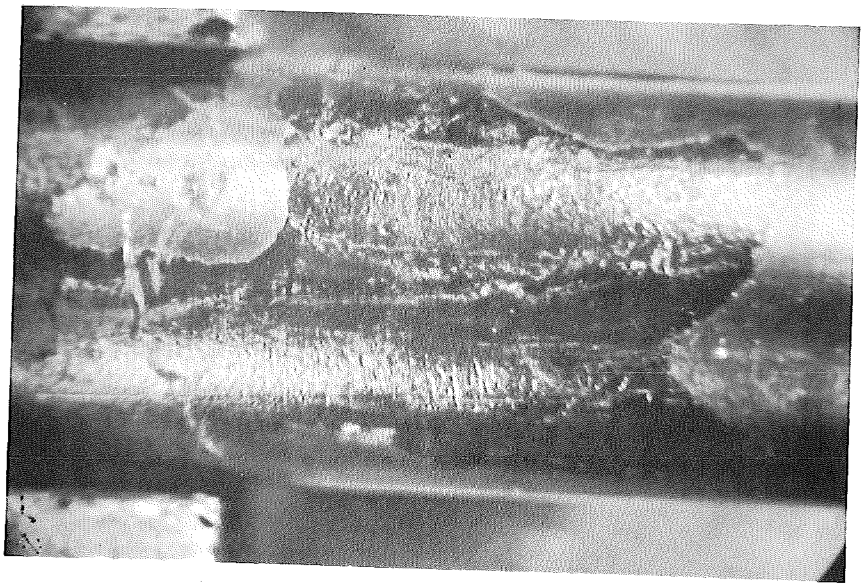


FIGURE 33. Portion of fuel element from Zr-feasibility Test, X-67
$T_s \sim 370°C$ $V = 9$ m/sec

3.2.3 First SAP Test X-58

The fuel charge for this test consisted of a single string of seven SAP-clad UO_2 fuel elements. Because of defects in the sheath, the irradiation was terminated after only eight hours. No significant fouling deposit was formed.

3.2.4 SAP Defect Test X-59

A single SAP-clad UO_2 fuel element with an intentional defect in the cladding was irradiated for 15 days. Gross failure of the cladding resulted. Heavy deposits formed on the exposed fuel pellets but only small amounts of fouling were observed on the sheaths themselves.

3.2.5 Finned Small Element Test X-708

Seven integral-finned SAP-clad UO_2 elements mounted end-to-end in a single string were irradiated for 720 hours. Two fin sizes and two coolant velocities were used in the test. The first four elements starting at the upstream or bottom end of the charge had short fins one mm high and a coolant velocity of 9.1 m/sec. The third element was located at the reactor flux centre line. The fifth element had full fins 2 mm high and a velocity of 9.1 m/sec. The sixth and seventh elements also had full fins but the coolant velocity was only 4.5 m/sec. Approximately one half of the seventh element projected above the D_2O level in the calandria.

The coolant had an iron content from 3 to 4 ppm, and a PCFT value of 75 mg.

After irradiation the first six elements had dark, relatively smooth films all of similar appearance except for the edges of the fins. The edges of the fins were shiny and free of deposits on the first, part of the second, and fifth elements. On the third, fourth and sixth elements the edges were covered with a dark deposit. The seventh element had a somewhat lighter deposit on the upstream portion but none on the edges of the fins, fading out to a negligible amount midway along the element, the position corresponding roughly to the D_2O level in the calandria. Figure 34 shows the upstream ends of the fifth, sixth and seventh elements. After preliminary examination six of the seven elements were stored under water. Unfortunately, this led to severe corrosion of the SAP sheaths which interefered with further examination. However, two layers could be detected in the deposits, an inner hard adherent layer less than 10 μm in thickness and an outer softer layer from 10 to 25 μm thick.

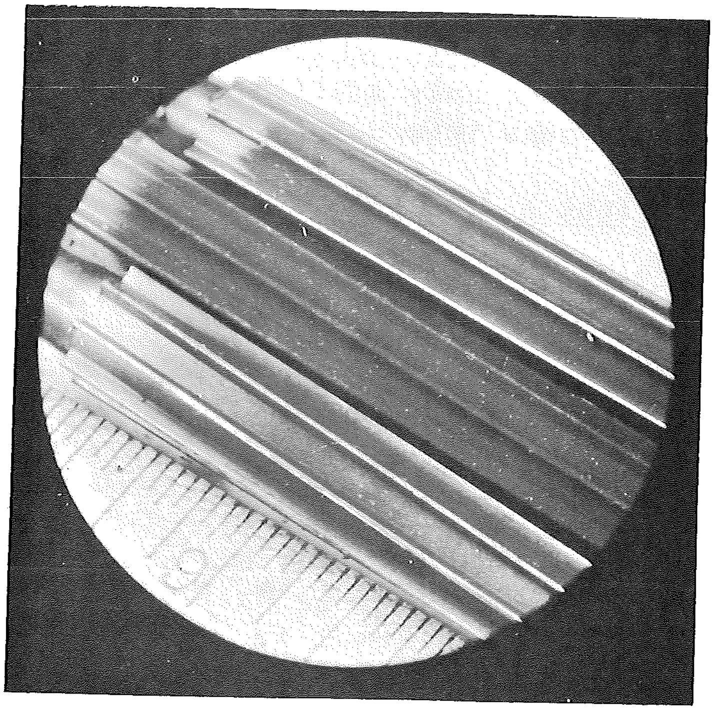


FIGURE 34. Upstream ends of 5th, 6th and 7th elements in X-708 Test.

Element	Coolant Velocity m/sec	Calculated Sheath Temperature, °C
5 (AHU)	9.1	443
6 (AHT)	9.1	435
7 (AHS)	4.5	404

3.2.6 First Trefoil Test - Phase 1 X-704-1

The fuel charge consisted of three trefoil bundles of 3 elements each. The elements in two of the bundles were SAP-clad UO_2 and in the third bundle were stainless-steel clad UO_2. The irradiation was terminated after 27 hours because of failure of one of the SAP-clad elements. The elements were spaced by spiral wire wraps.

Even though the exposure was short all nine elements were fouled. Visually the fouling of the SAP sheathed elements was indistinguishable from that of the stainless steel sheathed elements. Flaking of the deposits occurred on both types of elements apparently after the fuel was removed from the loop. The fouling was not visably different at positions near and away from the line of closest approach of neighbouring elements. Film thicknesses of 13 to 38 μm were measured. One film chip of unknown origin was almost 500 μm thick. X-ray diffraction analysis showed the presence of Fe_3O_4 and/or γFe_2O_3.

3.2.7 First Trefoil Test - Phase 2 - X-704-2

Phase 2 of the trefoil test consisted of two trefoil bundles, one SAP-clad and one stainless-steel clad. They operated for 31.5 days under conditions similar to Phase 1. Local conditions, however, were undoubtedly aggravated by severe distortion of the wire wrap on the elements.

Coolant analyses showed the water content, iron content and PCFT values decreased as the run progressed. Water contents ranged from 203 ppm to 126 ppm, iron contents from 6 ppm to 3 ppm and PCFT values from 81 mg to 38 mg.

The deposits formed on the fuel elements were more extensive and different in several ways to any previous deposits on X-7 fuel elements. The deposits upon examination in the Universal Cell were grossly flaked, to such an extent that it was not possible to relate completely film thickness to local areas. Film thicknesses ranged from less than 10 μm to greater than 1000 μm. The latter bridged the gap between fuel elements in areas where the flow was constricted by the distorted wire wrap. Fouling was present on all the end caps except those at the upstream end of the charge. The thinner areas of films were single-layered whereas the very thick deposits showed a stratified structure. The single-layered deposits had 53% residue on ignition, 30% iron and a density of 2.0 g/cm^3. X-ray diffraction detected the presence of Fe_3C plus traces of Fe_3O_4 and $Fe_{20}C_9$. The thicker deposits had 39% residue on ignition and contained alpha-iron and Fe_3C with no trace of oxide.

A number of specimens of film were sectioned and observed by metallographic techniques. These revealed a variety of structures. A sample of the thinner film showed predominantly the structure illustrated in Figure 35. However, on a few intermittent patches a second material was associated with it as in Figure 36. The samples of the thicker film were shown to be made up of a number of layers. In some, there were two distinct types of layers as in Figure 37. In others, the layers appeared to be all of a similar material as in Figure 38.

3.2.8 Fuel Element Fouling Test - X-706

This test was designed to investigate the effect on fouling of β-current, coolant velocity, and to a lesser extent surface temperature. Also, it would indicate by comparison with the parallel out-reactor Tests No. 3 and 4 the importance of radiation on the deposition process.

The fuel charge for this test consisted of seven Pu-Al fuelled stainless-steel sheathed elements mounted in a flow tube. A fifteen-fold difference in the β-current through the sheaths between various elements in each group was achieved by using two types of fuel cores. The I.D. of the flow tube was changed mid-way along the fuel string so that the upstream four elements had a coolant velocity of 2.5 m/sec, whereas, the downstream three elements had a velocity of 7.0 m/sec.

The coolant which had been in the X-7 loop for the X-704-2 test remained for use in the X-706 test. No special coolant treatment beyond the usual by-pass filtration through 5 μm sintered stainless steel filters was used. Analyses for the iron, water and gas contents and the PCFT values of the coolant during the test are reported in Table 13.

During the first part of the irradiation while the reactor was coming to equilibrium heavy-water level the coolant temperature was held at 260°C. As the moderator level neared equilibrium the coolant temperature was raised in steps until the conditions shown in Table 14 were reached.

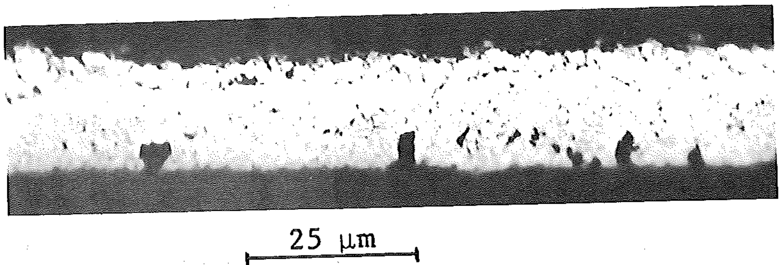


FIGURE 35. Cross Section of Film Flake Removed from Fuel Element in X-704-2 Test

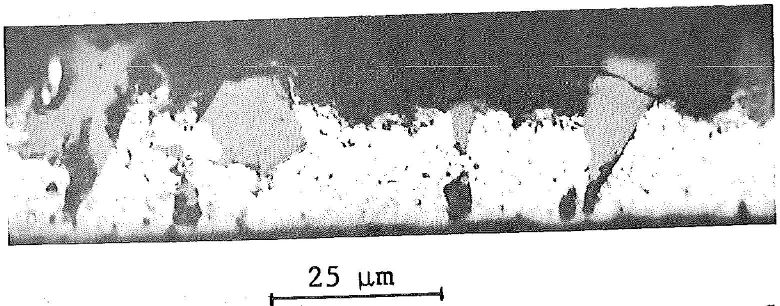


FIGURE 36. Cross Section of Another Portion of the Film Flake Shown in Figure 35.

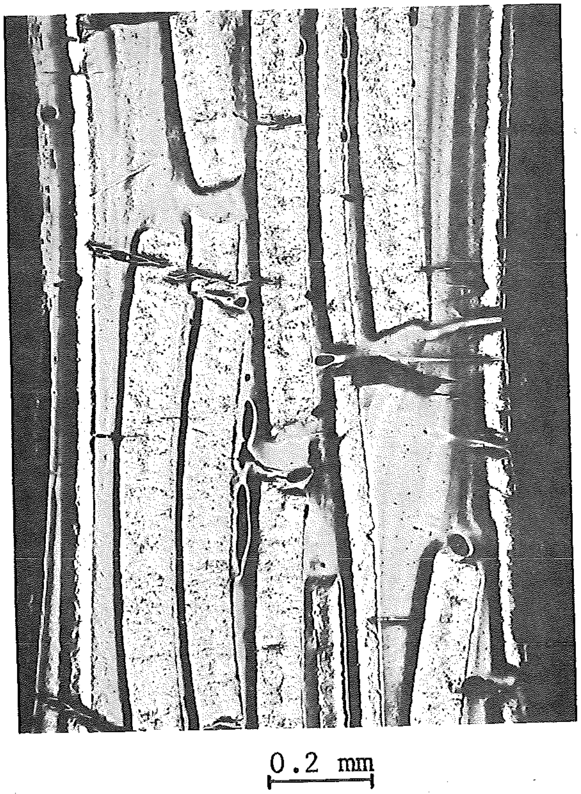


FIGURE 37. Cross Section of Deposit Removed from Throat between Fuel Elements in X-704-2 Test.

FIGURE 38. Cross Section of Deposit Removed from Fuel Element in X-704-2 Test.

The values given in this table are representative of 20.7 days of the 23.3 days of the irradiation.

The deposits on all the elements sharply outlined the fuel portion except on the upstream end of the upstream element where the start of the film was not sharp and was displaced a fraction of an inch into the fuelled portion. The interface at the centre of the element between the two fuel cores was indicated by a band of thinner deposit on several of the elements. The appearance of the film was quite different between the two groups of elements at different coolant velocities. However, within each group the appearance was similar. Deposits on the low velocity element were quite smooth and had a dull black lustre. The deposits on the high velocity elements were rougher, more adherent and had a sparkling appearance. There was no visual difference between the deposits on the low and high β-current elements. (See Figure 39).

The results of analyses and measurements of the film deposits are given in Table 14. In cross section the films on all the elements in the lower velocity sections showed similar structures as illustrated in Figure 40. The films on the three elements in the higher velocity regions were similar and were coarser-grained as illustrated in Figure 41.

During the test iron compounds were apparently transported from the loop surfaces to the fuel surfaces since the total iron content of all the films on the elements amounted to approximately 3300 mg, the equivalent of 10 ppm iron in the coolant. In comparison, the average iron content of the coolant during the test was 3 ppm.

3.2.9 Long-term Trefoil Test - Phase 1 - X-709-1

Since the Long-term Trefoil test, Phase 1, has been reported in detail elsewhere (11) only a brief description and summary of the results will be given here. The fuel for the test consisted of two trefoils each containing three SAP-clad UO_2 fuel elements about 60 cm long each. The cladding incorporated integral fins in which were mounted sheath thermocouples. The fuel was irradiated a total of 1819 hours over a period of slightly over three months.

Prior to the irradiation an adsorption column supplied by Atomics International was installed on a by-pass of the X-7 loop. The column was 53 cm in diameter and 46 cm long,

TABLE 13

X-706 COOLANT CONDITIONS

Time, Days	PCFT mg	Fe ppm	Water ppm	Gas Content cm^3/kg					
				H_2	N_2	CH_4	CO	Hydro-carbons	Total
0	74	-	-						
1	-	-	182						
2	91	-	-						
3	60	8	-						
4	-	-	-	13	168	8	1.8	31	224
6	83	3	-						
10	102	3	163						
11	-	-	-	22	206	19	0.9	41	289
13	103	-	-						
17	124	3	-						
18	-	-	-	59	107	10	1.6	-	-
20	78	5	-						
25	104	-	-						

TABLE 14

FUEL-ELEMENT FOULING TEST X-706

OPERATING CONDITIONS AND DEPOSIT OBSERVATIONS

Fuel Element	BJO	BJP	BJR	BJS	BJT	BJU	BJV
Velocity, m/sec	7.1	7.1	7.1	2.5	2.5	2.5	2.5
Reynolds no.	52,000	52,000	52,000	43,000	43,000	43,000	43,000
Pressure at outlet kg/cm^2	17.1	-	-	-	-	-	18.6
Bulk Temp, °C	356	353	350	348	347	347	346
Calculated surface Temp, °C	420	427	441	416	415	397	374
Av. heat flux, w/cm^2	65	76	93	25	25	19	10
Estimated beta current, µamp/cm^2	0.01	0.02	0.2	0.06	0.007	0.005	0.03
Duration, hr.	496						
FINAL DEPOSIT							
Thickness, µm average	46	50	30	25	20	20	20
min.	0	38	-	20	0	0	20
max.	51	51	-	38	25	30	20
Weight, mg/cm^2	15	16	21	13	7	5	5
µg/cm^2 hr	32	34	44	27	15	11	11
% loss on ignition	9	-	11	11	6	11	34
% Fe	58	-	58	62	68	58	37
% H	0.8	0.5	0.2	1.3	0.8	1.3	-
% C	5.9	5.0	7.3	15.7	12.9	14.2	-
Fe content of film, mg	600	640*	840	560	330	200	130
Fe compounds			oxide + trace αFe		oxide + trace αFe		oxide + trace αFe
Electrical resistance	low	-	low	-	-	low	-

Total = 3300 mg
= 10 ppm Fe in coolant.

* Assumed 58% Fe in deposit

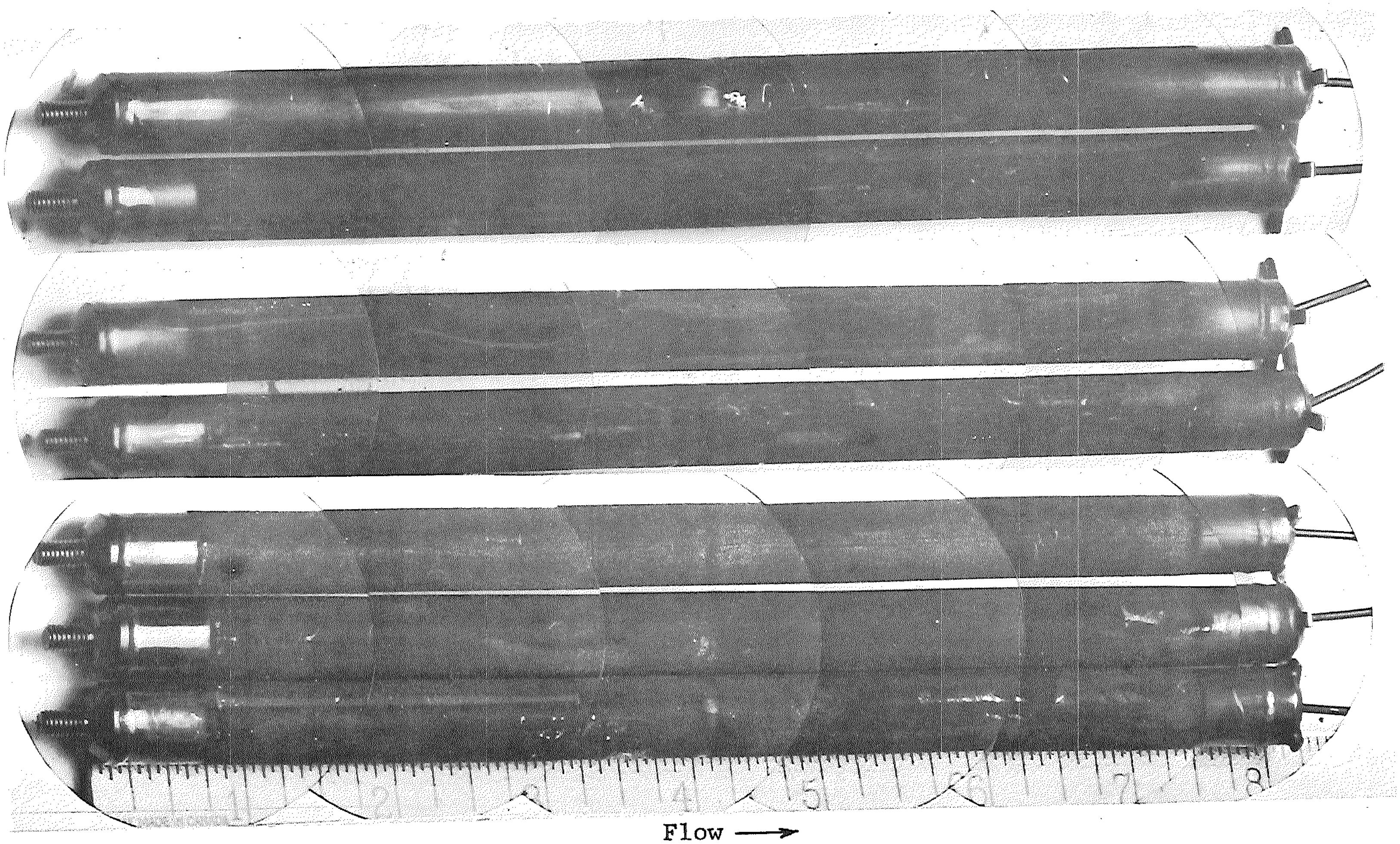


FIGURE 39. Fuel Elements from In-reactor Test X-706. (Scale in inches)

	Element	T_s, °C	V, m/sec	β, μamp/cm^2
(Top of photo)	BJV	374	2.5	0.03
	BJU	397	2.5	0.005
	BJT	415	2.5	0.007
	BJS	416	2.5	0.06

Element	T_s, °C	V, m/sec	β, μamp/cm^2
BJR	441	7.1	0.2
BJP	427	7.1	0.02
BJO	420	7.1	0.01
(Bottom of photo)			

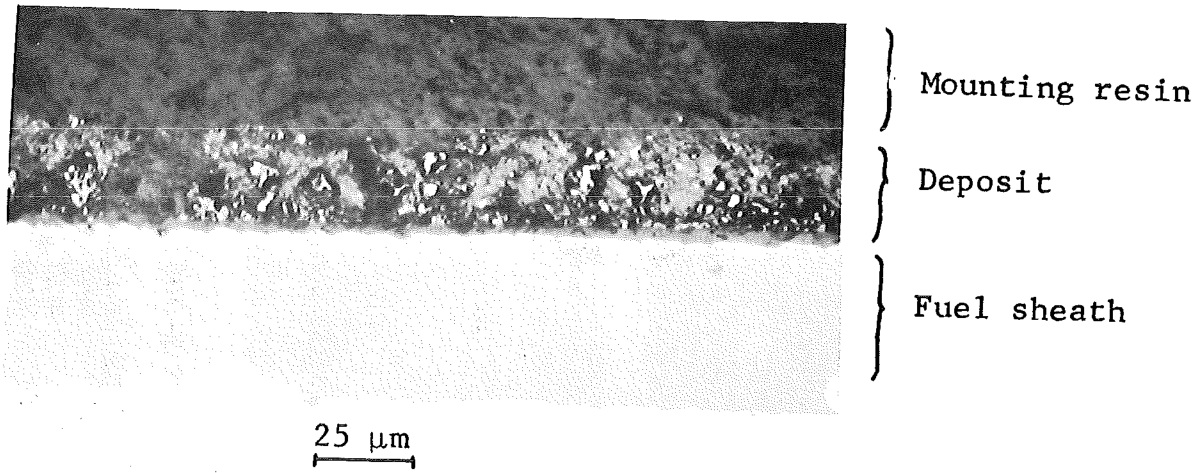


FIGURE 40. Cross section of deposit on element BJU in X-706 Test
T_s = 397°C; V = 2.5 m/sec

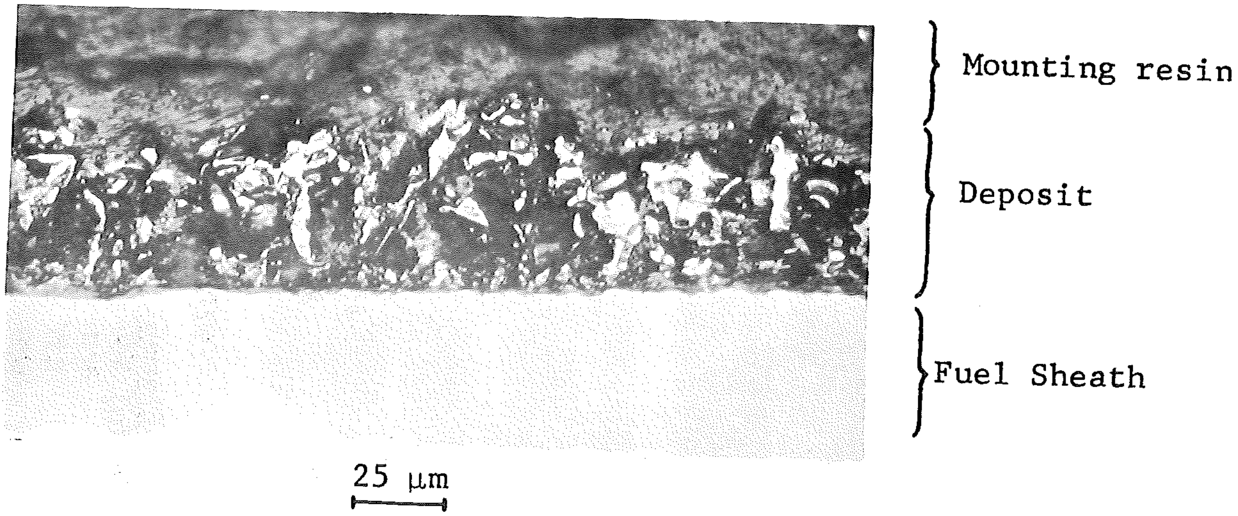


FIGURE 41. Cross section of deposit on element BJO in X-706 Test
T_s = 420°C; V = 7.1 m/sec

and contained Attapulgus clay which was dried in place before use. In order to clean up the coolant prior to installation of the fuel the coolant was circulated at a rate of approximately 20 l/hr through the column for about 2 weeks at an average temperature of 230°C. The clay was then replaced by a new charge and clean up continued at 270°C and at the same flow rate, for an additional two weeks before the fuel was installed. Operation of the column continued throughout the irradiation except for a few shut downs for repairs and modifications. The column operated 85% of the irradiation time. Coolant conditions varied somewhat during the test. The iron content ranged from less than 2 ppm to 23 ppm with an average of 3 ppm. Water contents ranged from 263 to 103 ppm with an average of 170 ppm. The PCFT values ranged from 1 mg to 51 mg, with an average of about 10 mg.

During the irradiation, the general trend was an increase in both sheath temperature and pressure drop. Over the total irradiation the sheath temperature increases were of the order of 60°C and the pressure drop increased from 3.0 to 4.3 kg/cm^2.

Upon examination, the heat transfer surfaces of the fuel were found to be covered with a smooth uniform film. The end caps at the inlet end of the inlet bundle were clean. All the other end caps were covered with film. There were no indications of heavy film formation at local areas in the bundle. The film thicknesses were measured both by microscopic examination of chips and by polished cross sections. The film thicknesses averaged about 40 μm at the inlet end of the inlet bundle, increased to about 75 μm by the middle of the charge and remained approximately constant at 80 μm throughout the length of the second bundle. No systematic circumferential variation in the film thickness was observed. X-ray diffraction analysis of the film showed mainly Fe_3O_4 and/or γ-Fe_2O_3 plus possible traces of alpha iron, FeC, γ FeO(OH). The film averaged 69% residue on ignition, 1.4% hydrogen, 32.7% carbon and 52% iron.

3.2.10 Beryllium Cladding Test - X-707

Ten UO_2 fuel elements of which eight were clad in beryllium and two in Zr-2 were irradiated as a single string in a flow tube as a cooperative test with the UKAEA and the French CEA.

Prior to this test the loop operated out-reactor for over a period of three weeks in order to replace the main pump and to test new test sections. For 130 hours of this period, the loop operated at temperatures over 425°C with some operation up to 445°C. The loop was returned to in-pile operation and make-up coolant added to dilute the high boiler which had been formed by pyrolysis during this testing period.

The low-flow large-diameter Atomics International column was replaced by a small-diameter high-flow clay column of AECL design in order to decrease the purification time constant and obviate the use of a preheater to maintain high bed temperatures. The new column was 15 cm in diameter by 80 cm long and operated at a flow of 165 litres/hour at 300°C. The first of these columns remained on stream for only ten hours during which period the iron content was reduced from 18 to 12 ppm and the PCFT value from 109 to 19 mg. After 18 hours on stream the second column had reduced the iron content to approximately 3 ppm, and the PCFT values to approximately 5 mg. These values were maintained throughout the remainder of the test by continued operation of the second column. During the first few days of the irradiation, the bulk inlet temperature was held to approximately 300°C until it became certain that the coolant was clean at which time the inlet temperature was raised to 355°C. The irradiation continued under these conditions for a period of two weeks. This corresponded to estimated sheath temperatures of from 430-460°C.

Post-irradiation examination showed the heat transfer surfaces to be covered with a very thin loosely-adherent deposit. When scrapping was found to be unsatisfactory, two other methods were used in an attempt to obtain film samples and film weights. One element (CAJ) was brushed with a fine bristle brush while immersed in carbon tetrachloride and the carbon tetrachloride subsequently evaporated to yield the film sample. The sample was 31% Fe, 6% Pb and had a residue on ignition of 60%. The weight was equivalent to a deposition rate of 0.2 $\mu g/cm^2$ hr. Element CAP was scrubbed with a wad of acetate staple soaked in carbon tetrachloride to remove all film. The dried staple was ignited to determine the residue after ashing. The result after correction for the ash content of the staple showed the total film from the element to yield 17.2 mg of ash. This equivalent to 0.2 mg/cm^2 or 0.6 $\mu g/cm^2$ hr based on the operating time at full temperature.

3.2.11 Uranium Carbide Test - X-711

In cooperation with Euratom eight uranium carbide fuel elements, six clad in SAP and two in s.s. were irradiated for 203 hours in the X-7 loop. During the first 100 hrs of irradiation the flow velocity was 8.2 m/sec and the average coolant temperature 306°C. Sheath temperatures ranged from 357-380°C. During the last half of the irradiation the coolant temperature was raised to 372°C., with a flow velocity of 8.4 m/sec. The sheath temperatures ranged from 415 to 438°C. During the irradiation the coolant had an iron content of 3 ppm or less and a water content of approximately 30 ppm. The PCFT values averaged about 4 mg.

The fuel charge exhibited sheath defects throughout the irradiation. The irradiation was terminated when a step increase in pressure drop across the fuel accompanied the reactor start-up. Post-irradiation examination revealed that one element had fractured into two parts. Very little deposit was observed on the fuel sheaths. The heaviest deposit was on stainless-steel sheathed element immediately downstream of the fractured element. (See Figure 42).

3.2.12 Long-term Trefoil Phase 2 - X-709-2

The fuel for this irradiation consisted of two trefoil bundles of SAP-clad UO2 fuel. Upon reactor start-up high activity in the coolant indicated at least one defect in the fuel and the irradiation was terminated after 19 hours. Because of this short irradiation time, the conditions were variable during the test. Approximate average conditions were a flow velocity of 7 m/sec, a coolant temperature of 295°C and sheath temperatures averaging about 400°C.

Prior to the installation of the fuel the loop had operated without a fuel charge of approximately a week and a half. During this time the third new six-inch clay column was installed. After installation of the fuel and before reactor start-up, the coolant was passed through a Molecular Sieve bed to further reduce the water content of the coolant. Coolant conditions during the test were iron content, less than 2 ppm; water content, approx 30 ppm; PCFT, 3 mg.

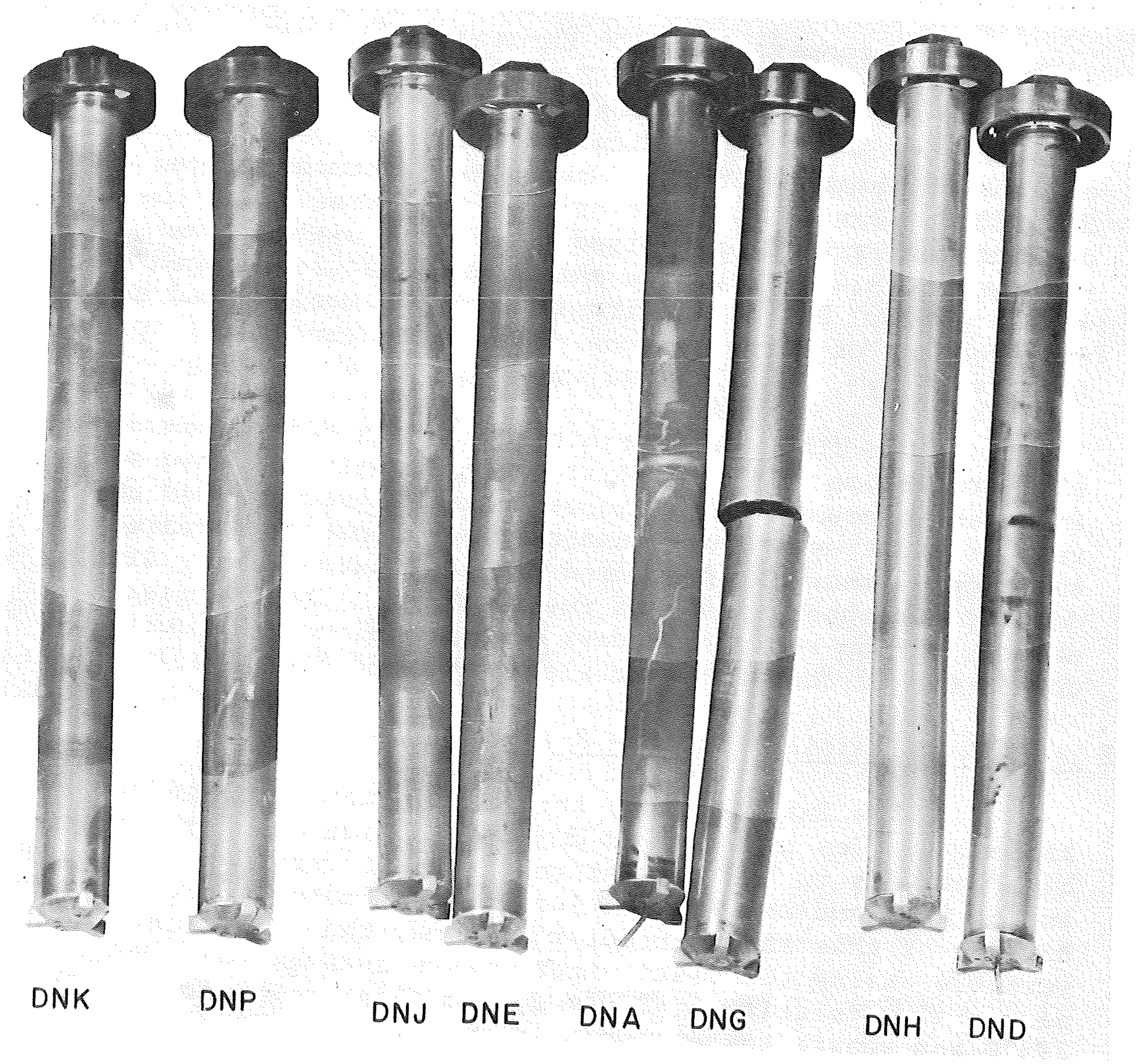


FIGURE 42. Irradiated elements from Test X-711 after rinsing, DND was upstream, DNK downstream in stringer.

Flow ⟶

Post-irradiation examination showed the heat transfer surfaces to be coated with a very thin loosely-adherent film. This was wiped from all the elements using the acetate staple technique used in the X-707 test. The total film from all six elements had a weight after ignition of 27 mg. This is equivalent to an ignited weight of 0.016 mg/cm^2 or 0.8 $\mu g/cm^2$ hr.

4. DISCUSSION OF RESULTS

4.1 Types of Fouling

The first impression from the experimental results is one of wide diversity in rates and types of fouling. Fouling rates varying over a 150-fold range under similar flow and temperature conditions have been experienced with basically similar organic coolants consisting of mixtures of ortho-and metra-terphenyl and 30% OMRE purification still bottoms. The deposited films ranged in composition from less than one to 95% organic material.

Detailed examination of the data suggests two distinct mechanisms of fouling, one of which involves deposits of basically inorganic material and the other of organic material. Because two separate processes are involved the discussion will first consider each separately, before looking at the combined result. Although both types of fouling can occur simultaneously, the presence of both organic and inorganic material in the deposited film does not mean that both mechanisms are active. For example, even though pure organic fouling is occurring inorganic material may be occluded in the depositing organic and thus be incorporated in the film. Also if a film is deposited by a pure inorganic mass transfer mechanism, some organic material may be detained long enough in the deposited film to be degraded and permanently held.

4.2 Mass Transfer of Inorganic Material

4.2.1 The Mechanism

The deposition of inorganic materials by a mechanism of mass transfer was first recognized in Out-Reactor Test No. 3. This test illustrated the main characteristics of this type of fouling.

(a) Iron was being transported from the walls of the system to the heat transfer surfaces of the test section by the coolant. The rate of release from the walls to the coolant was greatly accelerated when there was a high temperature surface in the system to act as a sink.

(b) The resultant deposit on the hot surfaces was predominantly inorganic material, mainly Fe_3O_4 and αFe.

(c) For equal surface temperatures greater deposition rates were observed at higher coolant velocities.

(d) The deposits were crystalline. At the higher coolant velocity Fe_3O_4 crystals up to 35 microns across with readily recognizable octahedral shape grew on the heat transfer surfaces. This produced a rough film of relatively high thermal conductivity which at least in the earlier stages tended to increase the over-all heat transfer coefficient.

The above characteristics suggest that the fouling layers were laid down by a mechanism involving low concentrations of iron in soluble form. Iron is believed to be extracted from the walls of the system into the coolant by a complexing agent. If the complex is less stable or less soluble at the high temperature of the surface than at the bulk temperature a driving force for mass transfer is available. The iron released precipitates and may deposit on the heat transfer surface as part of a growing crystal lattice. The remaining portion of the complex molecule is relatively stable and is carried off by the coolant stream to lower temperature regions where it again can complex with iron, and thus maintain the dissolved iron content of the coolant. The experimental evidence supporting this mechanism of fouling will be considered with reference to the four characteristics listed previously.

Definite evidence that iron was being transferred from the walls of the system to the heat transfer surfaces was observed in out-reactor Tests Nos. 3, 4, 6 and 9 and the in-reactor Test X-706. The ratios of the total iron found in the deposits of each of these tests to the amount of iron present in the total circulating coolant at any one time were 11, 13, 6, 10 and 3 respectively. Operation of the systems without any high temperature surfaces present had shown that once the iron content of the coolant had been reduced to about 2 ppm by purification the iron content of the coolant changed only very slowly. This was true whether or not purification of the coolant was continued. For example, before the start of test No. 9 the iron content of the coolant changed at the most 2 ppm during a period of 220 hours. The iron content during the 44 hours which the test sections were heated remained constant at 3 ppm. However, during this same period iron equivalent to 20 ppm in the total coolant charge was deposited on the test sections. Since the concentration of iron did not change when the power to the test sections was on, the rate of release from the walls of the system must have increased to match the rate of its removal by the test sections. These observations are akin to those which would be expected if a mechanism involving soluble iron were acting. The solubility of the iron must be the result of an impurity present in the loop in limited concentrations such as a complexing agent. A simple solubility of iron or iron oxide in the major coolant components without the aid of a complexing agent cannot be responsible, otherwise, this behaviour would have been encountered to approximately the same extent in all the tests.

To account for the rate of mass transfer the soluble iron concentration need only be very small. Even at the rapid rates of mass transfer in Test No. 9 the change in iron concentration in the coolant as it passed once through the test sections was only 0.013 ppm.

It is quite possible that most of the 3 ppm of iron in the organic was not present in a soluble form but rather as suspended particulates which contributed at most a minor portion of the deposits.

The deposits in tests 3 to 9 were predominantly inorganic material. Based on the carbon and hydrogen analyses the films ranged from 53 to 99% inorganic with an average of around 90% inorganic material. Carbon and hydrogen analyses of the in-reactor deposits are not complete, but based on the residue on ignition the in-reactor deposits were from 53 to 90% inorganic except for the X-704 test in which a value of 30% was obtained. The complexing agent is almost certainly organic. Since films with very low organic contents can be formed by the mass-transfer mechanism apparently the complexing agent is not deposited simultaneously.

The inorganic portions of the deposits were predominantly iron or magnetite. It is not possible from the experimental evidence available to distinguish whether or not the iron was carried to the surface in association with oxygen. In the films from the out-reactor tests α-iron and magnetite were observed both separately and together. In the films from the in-reactor tests only oxides were detected in quantity, except in test X-704-2 and X-706. In the former the iron was present almost exclusively as Fe_3C and in the latter some α-iron was also present.

In the out-reactor Test 3, 4, 5 and 6 and in the X-706 test in which surfaces were exposed at two coolant velocities higher deposition rates were observed at the higher velocities. The change in deposition rate was proportional to between the one-half and the first power of velocity. The velocity dependence is similar to that for mass transfer processes between a turbulent fluid stream and a solid interface.

In the tests in which there were two coolant velocities not only was there a difference in deposition rate between the regions of different velocity but also a marked difference in the appearance. The deposits in the high-velocity regions were noticeably rougher and more sparkly than in the low-velocity regions. In Test No. 9 and in the higher velocity portions of tests 3, 4 and 6 there were readily recognizable crystal faces which were equilateral triangles up to 35 microns across. A few complete octahedral crystals were identified. This corresponds to the normal crystal habit of magnetite. It must be concluded that these crystals grew in place on the heat transfer surfaces. It is highly unlikely that such large particles could be formed elsewhere in the system and be carried in the coolant stream without being removed by the purification system. Even if they were carried by the coolant the smooth test surfaces with the highest coolant velocity in the system were the most unlikely places for their deposition. Since the deposition involves crystal growth, transport of the iron in solution is inferred. The previous three characteristics of the film could be satisfied equally well by a mechanism involving colloidal type transport with only a small concentration of stabilizing agent present. However, the growth of crystals denies the colloidal-type mechanism.

4.2.2 Effect of Surface Temperature

Deposition rates in the out-reactor Tests 3 to 9 in which the films were predominantly inorganic are plotted against initial surface temperature in Figure 43. Rigorously, to obtain the effect of surface temperature on mass-transfer type fouling, the organic fraction of the deposits should not be included. Unfortunately the lack of carbon and hydrogen analysis for some of the films prevents this correction. However, since in nearly all cases the correction is small, the conclusion is not significantly affected. The data clearly indicate that the deposition rate is increased by increased surface temperatures and can be approximated by an exponential relation. The exponential relation best representing all the data predicts a ten-fold increase in deposition rate for a 90°C increase in surface temperature (or a doubling for a 27°C increase).

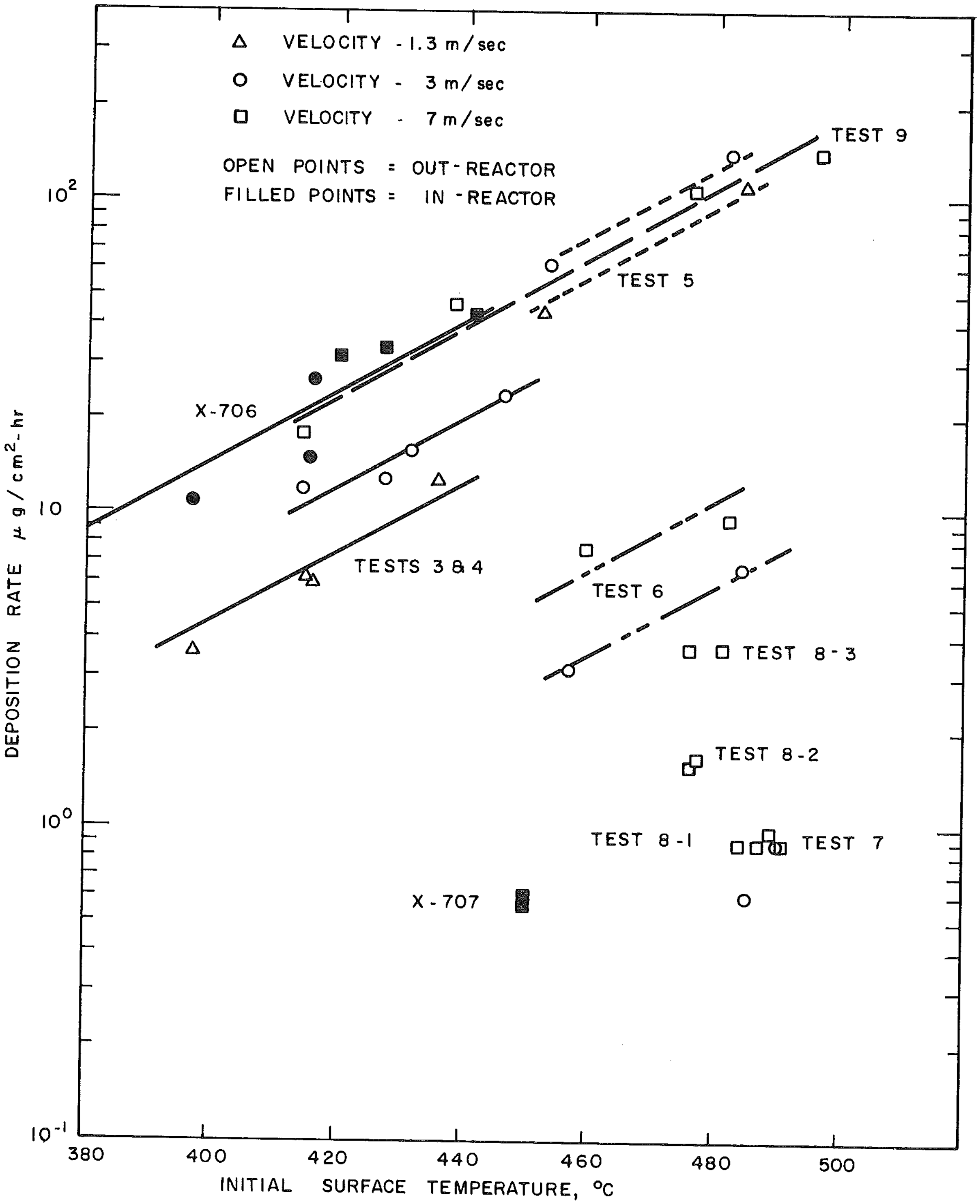
It has been assumed that surface temperature is the pertinent parameter. Since there was little variation in bulk temperature from test to test, the temperature difference between coolant and surface could be used equally well for correlation. Which is the more important parameter cannot be defined from the present data. However, the proposed mass-transfer mechanism implies dependence on a driving force which results from a change in stability or solubility complex with temperature. This driving force is likely to be a function of both temperature level and temperature difference.

4.2.3 Effect of Velocity

In each of out-reactor Tests 3, 4, 5, and 6 a pair of test sections were used in which the coolant velocities differed by a factor of 2.35. At equal surface temperatures, the deposition rates in the sections with the higher velocities were from 1.4 to 2 times higher than in those with the lower velocities. This is equivalent to a functional dependence of deposition rate on velocity raised to a power in the range of about one half to less than one.

As was the situation with surface temperature, the data do not allow the separation of the effects of two possible parameters. Both velocity and Reynold's number differed in the test section pairs. The similarity of related mass transfer processes would favour the Reynold's

FIGURE 43. DEPOSITION RATES FOR ORGANIC COOLANT WITH VARIOUS CLEANUP METHODS



number as the better parameter. In the X-706 test in which the channel shape was annular, the velocity differed by a factor of 2.84 for two groups of elements whereas the ratio of Reynold's number was only 1.2. The effect on the relative deposition rates was inconclusive because of experimental scatter. However, since the difference in crystal size and appearance of the films on the two groups was equally as great as in the parallel out-reactor Tests 3 and 4 where the velocity ratio was 2.35 and the ratio of the Reynold's numbers was 1.6, velocity appears to be the more important parameter. Further experiments would be necessary to establish the correct parameter with certainty.

4.2.4 Effect of Radiation

Radiation could affect fouling rates in two ways; by causing chemical changes in the bulk coolant and by contributing in the mechanism of the actual deposition process at the heat-transfer surfaces. Since the X-7 loop has a very large ratio of coolant volume out-reactor to in-reactor (~300:1), radiation-induced chemical changes in the bulk coolant are diluted to such an extent as to be masked by small variations in the make-up to the system. The effects on fouling of radiation damage to the coolant are being studied in other equipment and will not be reported here. Tests in the X-7 loop do, however, provide information on radiation effects at the heat-transfer surfaces.

As is evident in Figure 39 the surfaces of the end caps of fuel elements were frequently almost free of deposits even when relatively heavy films formed on the heat-transfer surfaces. Since the intensity of the radiation, both gamma ray and neutron, is very similar at both types of surface, radiation does not have a dominant effect on the deposition process.

The specific effect of beta current through the sheath hypothesized by Parkins (4) was not observed in the X-706 test. Since beta currents were deliberately varied from element to element over the range 0.005 to 0.2 μamp/cm^2 and no effect on fouling was detected, beta current was not a major parameter.

Comparison of the results of the out-reactor Tests 3 and 4 with the in-reactor Test X-706 as plotted in Figure 43 shows the effect of radiation on the deposition process to be not large, probably less than a factor of two for the NRX radiation at a surface temperature of about 420°C.

Further less-positive evidence of the effect of radiation comes from comparison of the in-reactor X-707 test and the out-reactor test No. 7. The coolant conditions for the two tests should be similar on the basis of treatment and impurity analyses. If allowance is made for the differences in surface temperatures and coolant velocity, the radiation effect on the deposition rate appears to be less than a factor of two and certainly less than a factor of five.

4.2.5 Effect of Coolant Composition

The major components of the coolant, the polyphenyls, do not cause significant fouling by mass-transfer mechanisms. The concentrations of biphenyl, terphenyls and high boilers were similar in the coolants used in all the tests, both in-reactor and out-reactor, yet fouling rates varied over a range of over two orders of magnitude. There is nothing to indicate that fouling rates even lower than those attained in out-reactor Test No. 7 cannot be achieved with these same concentrations of polyphenyls.

The large changes in fouling rate must, therefore, be attributed to changes in the concentration of impurities present in the coolant in small amounts. The composition of the coolant with respect to impurities is the most important factor in determining the fouling rate on a hot surface cooled by a polyphenyl coolant. This applies to the mass-transfer type of fouling and also, as will be discussed later, to the organic-type fouling.

Reference to Figure 43 illustrates the changes in the fouling tendency as more efficient methods of impurity removal, or "cleanup", are used. Out-reactor Tests 3 and 4 are examples of fouling rates with coolant from which most of the suspended material larger than 5 microns had been removed by filtering. Test No. 6 illustrates the reduction in fouling rate obtained by a once-through contacting of the coolant through a bed of

Attapulgus clay. A more thorough cleanup of the coolant in Test No. 7 by continuously recycling a part of the loop flow through a clay bed as well as reducing the water content of the coolant resulted in a further large decrease in the fouling rate. A similar series of decreases in fouling rate as the efficiency of cleanup was increased can be followed in the X-7 tests.

In out-reactor Test No. 9, a very high fouling rate was observed even though the coolant was continuously treated with Attapulgus clay. Presumably the particular types of impurities removed from the coolant by the clay in Tests No. 6 and 7 were again removed in Test No. 9. However, an additional impurity, later shown to be chlorine, which was not effectively removed by the cleanup methods used was capable of promoting rapid fouling of the mass-transfer type. There are, therefore, more than one type of impurity which can cause mass-transfer. The attainment of minimum fouling rates must involve the removal of all impurities, where "impurities" are defined as those materials which when present in small concentrations promote fouling. Those which promote inorganic fouling do so by increasing the solubility of iron or other metal compounds in the coolant.

No concerted effort was made to identify the specific impurities which increased the iron solubility in these studies. Chlorine is the only one positively identified. Analysis showed that chlorine had concentrated in the 9-3B film to the extent of 250 ppm. Because of the potency of chlorine in promoting mass-transfer fouling a program of experiments is underway to determine the chemical form in which it participates, and the best method of removing it from the coolant.

4.3 Organic-type Fouling

4.3.1 Mechanism

The characteristics of the organic type of fouling are:-

(a) Deposition rates are increased by increased surface temperatures and decreased coolant velocities.

(b) The deposits exhibit an inlet effect and in some instances a downstream-end effect.

(c) The film composition is similar to the composition of the particulates in the coolant.

Since fouling of predominantly the organic type was observed only in out-reactor Tests 1 and 2 in this study the data obtained on this type of fouling are much less complete. A few of the other tests had films of sufficient organic content to provide additional observations. Characteristics (a) and (b) are illustrated by the data. A greater source of data is OMRE in which organic-type has predominated. Characteristic (c) is based on these data. The OMRE and other data has been discussed by Gercke (5) and a mechanism proposed. The basic mechanism depicts the organic type of fouling as a deposition on the heat-transfer surfaces of particulates of micron and submicron size from the coolant. A fraction of the particles which impinge on the surface by diffusion are chemically bonded to it, the fraction being governed by a sticking probability. The sticking probability is a function of the chemical reaction rate, the residence time, the coolant velocity and the particle size.

The observations from the studies reported here do not disagree with this basic mechanism. However, the detailed interpretation and the rating of the relative importance of the parameters are different in several important aspects as discussed in the sections following.

4.3.2 Effect of Surface Temperature

The results of the out-reactor Tests 1 and 2 demonstrate the increase of fouling rates by increased surface temperatures. The magnitude of the effect is approximately a doubling of the deposition rate by a surface-temperature rise of 30°C. This compares with doubling intervals of 11° and 41°C illustrated by Gercke for out-reactor tests.

Gercke concluded that even though the effect of surface temperature was large under out-reactor conditions, it was insignificant in a reactor radiation field because of the over-riding effect of the radiation. Although no accurate data are available from the X-7 tests to directly evaluate the in-reactor effect of surface temperature on organic-type fouling, the radiation effect found was too small to substantiate Gercke's conclusion as will be discussed in Section 4.3.4.

4.3.3 Effect of Velocity

Tests 1 and 2 also show that organic fouling rates decrease with increasing velocity. Additional evidence in Table 15 obtained from Tests 3, 4 and 6 using the film analyses and total deposition rates to obtain the deposition rate of organic material, also illustrate this effect. Without exception the rates were significantly less at the higher velocities, the average reduction being a factor of 1.9 for velocity increases of a factor of 2.3. This is, however, a much less sensitive dependence on velocity than that indicated by the Atomics International results. The AI data show a factor of 300 decrease in rate for a velocity increase of a factor of 2.8.

The values reported here were obtained from pairs of surfaces exposed concurrently to the same coolant at different velocities. They do not, therefore, include the large uncertainty present in the AI data which were obtained with only one velocity present in any particular test and without precise control of coolant conditions from test to test.

The in-reactor data in Table 15 from the film analyses and deposition rates in the X-706 test do not show a large influence of coolant velocity.

4.3.4 Effect of Radiation

The effect of radiation on the deposition of organic type films (excluding its effect on the bulk coolant as in Section 4.2.4) was not large. For example, in the X-706 test the organic deposition rates ranged from 1.7 to 4.6 $\mu g/cm^2$ hr compared to a range of 0.6 to 6.1 in the corresponding out-reactor tests. Also, in the X-7 tests the fuel element end caps, which saw essentially the same radiation field as the remainder of the fuel can, always collected less film and were frequently almost free of fouling.

The minor effect of radiation on fouling in X-7 is contrary to the observations of Gercke who concluded that in-reactor fouling rates were several orders of magnitude higher

TABLE 15

DEPOSITION RATES OF ORGANIC MATERIAL

Test Section or Fuel Element	Velocity m/sec	Surf. Temp. °C	Total Deposition Rate $\mu g/cm^2$ hr	% (C+H) in deposit	% Loss on Ignition	Organic Deposition Rate, $\mu g/cm^2$ hr
Out-reactor Tests						
2-2B	3.8	480	2.5	-	95	2.4
2-3B	1.7	485	4.3	-	91	3.9
3-2B	1.3	416	6.1	14.4	-	0.9
3-3B	3.0	431	16	4.0	-	0.6
4-2B	1.3	436	13	47.0	-	6.1
4-3B	3.0	446	24	18.2	-	4.4
6-2B	3.1	484	6.8	12.5	-	0.8
6-3B	7.2	482	9.6	3.0	-	0.3
X-706 Test						
BJO	7.1	420	32	6.7	-	2.1
BJP	7.1	427	34	5.6	-	1.9
BJR	7.1	441	44	8.5	-	3.8
BJS	2.5	416	27	17.0	-	4.6
BJT	2.5	415	15	13.7	-	2.1
BJU	2.5	397	11	15.5	-	1.7
BJV	2.5	374	11	-	37	4.1

than those experienced out-reactor. The difference between the two conclusions can only partly be reconciled by the differences in experimental conditions. Sheath temperatures in X-7 have been in general considerably higher than in OMRE, i.e. 420° to 470°C compared to 400°C. The energy deposition rate in the coolant next to the sheath has been lower in X-7 than in OMRE by a factor of about 3 for the trefoils and about 6 for the single elements. If it is assumed for purposes of comparison that temperature and radiation-induced fouling are independent and additive, then an environment of higher energy deposition rates and lower surface temperatures would be expected to show a greater apparent effect of radiation on fouling than one with the reverse of these conditions. If, in X-7 at a surface temperature of 430°C on a single element the radiation doubles the fouling rate compared to the out-reactor rate, the temperature and radiation-induced rates are equal. Transposing to OMRE conditions increases the component from radiation by a factor of 6, and reduces the temperature component by a factor of 2. By this reasoning if the ratio of fouling rates in-reactor to out-reactor in X-7 is 2 then at OMRE conditions it would be 13. This is a much smaller effect of radiation than that deduced by Gercke.

4.3.5 Effect of Heated Length

It was first noted in the X-704-2 Test that the element end caps at the upstream end of the fuel charge were clean and the other three sets further downstream were somewhat fouled. The same observation was repeated in several subsequent tests. In the X-706 Test the start of the fouling on the upstream end of the upstream element was delayed about 0.5 cm compared to the other six elements.

In out-reactor Tests 3, 4, 5 and 6 the electrical resistance of the fouling films were observed to increase over several orders of magnitude with distance downstream from the start of the heated surface. The weight per unit area of deposit also increased with increased downstream distance, the major portion of the change occurring in the first 5 cm.

All these observations indicate that the chemistry of the coolant underwent changes significant to fouling when small portions of it were heated to a temperature above that of the bulk of the coolant. A steady state was reached in a very short time. The change did not decay away in the interval between test sections or fuel elements in line but had disappeared in the time required to make one circuit of the loops.

Further study of this phenomenon might provide useful insight into the fouling mechanism. However, no effort has been devoted to this aspect in the present study.

4.3.6 Effect of Coolant Composition

Much of the discussion in Section 4.2.5 as to the effect of coolant composition on the mass-transfer type of fouling applies equally well to organic fouling. Organic fouling rates varying over a range of more than two orders of magnitude were observed in the out-reactor tests. The variation must be almost all attributable to differences in the impurity concentration in the coolant, the most important parameter affecting the fouling rate.

Gercke (5) has outlined the mechanism of organic-type fouling and presented evidence on the source and structure of the particulates. The data of the present studies permits some further elaboration. In Test No. 2 a film which was 95% by weight organic material was deposited. Since this is equivalent to 99% by volume it is apparent that many of the particulates were entirely organic.

Organic-type fouling rates can be reduced to low levels by treatment of the coolant with Attapulgus clay. The material removed by the clay has not been identified. Analysis of the film, of course, is of little help since the deposit has undoubtedly been degraded considerably during the prolonged period at the high surface temperatures.

4.4 Control of Fouling in Organic-cooled Reactor Systems

Since the findings from these experimental studies should provide information for reactor design, the results must be examined with this end use in view.

The deposition of a fouling layer on the heat-transfer surfaces of the fuel elements is detrimental to the operation of the reactor since it increases the resistance to heat transfer and increases the pressure drop across the core. The amount of deposit which can be tolerated from the heat-transfer point of view in an economic reactor is so small (approximately

25 μm layer) that the corresponding effect on the pressure drop is not likely to be significant. The rates observed in the out-reactor Test No. 7 and the in-reactor X-707 Test are the only ones of those reported here which are approaching values permissible in a reactor having the high heat fluxes required in the OCDR concept.

Since the reactor condition will involve only thin fouling films, the hazards of flaking deposits blocking narrow coolant channels as in Core II of OMRE and in the X-704-2 Test are not likely to exist. No evidence that flaking occurred during operation has been observed in the X-7 or out-reactor tests with films less than 50 μm (0.002 in) thick. If there is no film flaking and the reactor system is at steady state the consideration that film buildup is linear with time is a workable assumption.

The results presented have been expressed as film weights per unit time (μg/cm^2 hr) whereas the criterion for the reactor must be based on heat-transfer effects. It is apparent from the results that a general relation between the two is not possible; that is the density and effective thermal conductivities of the deposits varied greatly. For example, the effective thermal conductivities of the films on test sections 2-3B and 6-2B were, respectively, 0.009 and 0.003 w/cm °C (0.5 and 0.2 BTU/hr ft °F) and on test section 9-3B was a negative value. Based on heat transfer alone the fouling in Test No. 9 is acceptable for a reactor since it resulted in a drop in surface temperature rather than a rise. However, it is obvious from the deposition rates on a weight basis that is is not acceptable on the long term. Because of the inability to predict long term heat-transfer effects at low fouling rates from measured rates in short tests the approach in the AECL studies has been to use the measured deposition rates in short tests to study the fouling mechanisms and the effectiveness of coolant cleanup methods. Once a cleanup method has been proven a long-term out-reactor test will be run to determine the heat-transfer effects accurately at low fouling rates. Long-term in-reactor tests will provide less accurate data but in a reactor environment. The present target for an acceptable deposition rate is less than 25 μm/yr (0.001 in/yr) or approximately 0.3 μg/cm^2 hr.

The experimental studies have indicated a number of parameters which, theoretically at least, could be varied by reactor design to minimize fouling. The major parameters are surface temperature, coolant velocity, and coolant composition.

It has been shown that coolant composition can cause effects which are at least an order of magnitude greater than those from the other parameters and it is from this parameter that most of the control of fouling must come. There is little incentive to decrease surface temperatures, or limit coolant velocities, for example, in order to lower the fouling rate. (It is still necessary, of course, to allow for the effect of fouling in setting the maximum sheath temperature).

Present experience indicates that satisfactory control of fouling rates can be obtained through control of the impurity content of the coolant. Substances known or suspected to be detrimental such as chlorine, oxygen, lead and water are best controlled by excluding them from the system. The small amounts of them which do enter can be removed along with the products of coolant degradation and system corrosion to satisfactory concentration levels by the coolant purification systems, i.e. the Attapulgus clay columns and filters, the chlorine-removal column, the degassing system and the distillation system.

In order to follow the effectiveness of the purification systems some method of evaluating the fouling tendency of the coolant is needed. The best overall method available is the Small Probe Fouling Test (12) in which a sidestream of reactor coolant is passed over an element electrically heated to 480°C at a velocity of 3 m/sec and the weight of the deposited film determined. This is in essence a small-scale duplication of conditions representative of the fuel element surfaces except for the lack of the radiation field. Since radiation has been shown to have only a relatively small effect on the deposition rate this is not a serious difference. The method is sensitive to fouling of both types. Unfortunately, a relatively long test (at least a week) is required to give a quantitive value for a clean coolant. Increasing the surface temperature by say 100°C is not a satisfactory way of accelerating the test since it would introduce boiling and might also lead to coking or other non-representative deposition.

Other analyses are helpful for more rapid detection of trends and for indicating sources of coolant contamination. Analyses for the concentration in the coolant of iron and chlorine and the residue on ashing are indicative of the order of magnitude of the fouling rate. The experimental organic-type fouling rates were not correlated by the PCFT values.

PCFT values of about 10 mg or greater indicate that organic fouling rates are prohibitively high. However, low PCFT values apparently do not guarantee that the coolant is satisfactory. The percentage of benzene insolubles also has not been correlated with fouling tendency.

5. CONCLUSIONS

1. Fouling films are deposited on heat-transfer surfaces in organic-cooled systems by two mechanisms, one involving impurities soluble in the circulating coolant, the other involving impurities insoluble in the coolant. The first is a mass-transfer type of mechanism in which iron from the mild steel piping is taken into solution and deposited as a result of the decreased solubility or stability of the iron compound at the higher temperatures existing at the heat-transfer surfaces. Deposition by the second mechanism occurs as a result of the chemical bonding of the organic portion of particulate material in the coolant to the heat-transfer surface. Since the mass-transfer mechanism leads to predominantly inorganic deposits, whereas the particulate mechanism leads to predominantly organic deposits, large variations in the composition and structure of fouling films are encountered.

2. The rate of fouling by both mechanisms is mainly controlled by the impurity content of the coolant, the important impurities being those which increase the dissolved iron concentration and those organic components which are insoluble in the coolant. Insoluble inorganic material suspended in the coolant will enhance the effect of the latter. A terphenyl-based coolant free of these impurities will not result in significant fouling under conditions pertinent to reactor operation.

3. Coolant velocity affects the rate of fouling by the two mechanisms differently. Increased velocities increase the deposition rate by the mass-transfer mechanism but decrease the rate by the particulate mechanism. For both mechanisms the change in rate is proportional to no more than the first power of the velocity.

4. The higher the temperature of the heat-transfer surface the higher the deposition rate by both mechanisms. Approximately, the effect is a doubling of the fouling rate for each increase of 30°C.

5. Nuclear radiation does not cause an important increase in the deposition rate. The relative magnitude of the effect on the deposition process depends on the energy absorption rates and surface temperatures involved. However, at the conditions expected in organic-cooled heavy-water-moderated power reactors radiation will not be a decisive factor in determining the deposition rate.

6. Continuous coolant cleanup through Attapulgus clay, combined with the exclusion of such contaminants as chlorine, has been shown to reduce deposition rates under representative reactor conditions to 1 $\mu g/cm^2$ hr. There is no reason to believe that further purification could not reduce rates to 0.3 $\mu g/cm^2$ hr, an acceptable level in a power reactor.

6. ACKNOWLEDGEMENTS

Many people in the Chemistry and Metallurgy and the Operations Divisions contributed to the studies reported here. M. Rainville operated the 250-0-1 loop including the assembly of the test sections and the data processing.

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