

# Heat-Transfer Studies on Some Stable Organic Fluids in a Forced Convection Loop

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STABLE organic fluids used as coolant moderators have been under study for a number of years (3, 11); physical, chemical, nuclear, and engineering properties of many of these fluids, relative to this use, have been investigated (1, 6, 18, 22). Even though these fluids, compared to water, have poorer heat transfer properties and stabilities, their relative inertness to container materials, their good nuclear properties, and their high boiling temperatures make them attractive for many applications.

A more precise measurement of film coefficients and a study of possible fouling of heat-transfer surfaces caused by pyrolytic degradation of the fluids were indicated by the in-pile loop experiments of McEwen (13). At U. S. Naval Research Laboratory (NRL) a forced convection, out-of-pile loop was used to study heat-transfer properties. Special emphasis was placed on film coefficient measurements and on possible fouling of heat-transfer surfaces. The system was designed to facilitate these studies at heat-transfer surface temperatures to 850° F., with heat fluxes approaching 500,000 B.t.u./hr. sq. ft., and with fluid velocities approaching 50 feet/sec.

The film coefficients measured for seven fluids have been correlated by an equation of the Dittus-Boelter type, and this equation is recommended for the estimation of film coefficients of polyphenyl and aliphatic oils for specified conditions of flow and temperatures.

## EQUIPMENT

The heat-transfer loop constructed for this study and described previously (19) consisted essentially of pump, a flow-control system, a preheater unit, a gas-separator unit, a heat sink, a surge tank, and a make-up tank. Low-carbon steel pipe (welded) was used in the system with the exception of the test section which was made of Stainless steel, Type 347. Design pressure was 500 p.s.i. at 600° F. Fluid was circulated by a Byron-Jackson, single-stage, centrifugal pump or by an Aurora, two-stage, turbine-type, centrifugal pump. Flow through the system was controlled at desired constant rates between 1.5 and 15 gal./min. by a throttling motor valve that was actuated by a variable-area, Fischer-Porter flowmeter and controller. Desired bulk-fluid temperatures at the entrance of the test section were accurately maintained by an annular-type, single-pass, preheater unit equipped with external electrical heaters. Provision was made at the gas separator so that liquid and gas samples could be collected during operation of the loop. A single-pass, annular-type, heat exchanger, in which a controlled amount of water was flashed to steam at atmospheric pressure, was used as a heat sink. The surge and make-up tanks were arranged in the system to permit evacuation, filling, pressurization (with argon), and maintenance of liquid levels.

**Test Section.** The electrically heated test section was a horizontal, single-pass exchanger in which the coefficients of heat transfer were determined over the central 12 inches. A careful analysis and design of the test section was made to minimize any error in coefficient measurements which could result from entrance effects, heat losses, and control variables. A calming length with a length to diameter ratio of 25 preceded the measurement section. Calculations with entrance region equations and observed axial temperature

profiles indicated negligible entrance region effects. Axial conduction of heat and variation in generation of heat because of change in electrical resistance along the tube wall were insignificant.

The test-section tubes (Table I), 24 inches in length, were flanged on both ends to mixing pots. One of the flanges was electrically insulated and the other grounded. Alternating current, 60-cycle, was passed through the center 18 inches of the tube between silver busses; and power input to the measurement section, the central 12 inches of tube, was measured between voltage taps. Temperatures were measured at nine positions along the outer surface of the measurement section with a thermocouple system patterned after those used for precision thermal conductivity studies at this laboratory. Accuracy of temperature measurements with stabilized, calibrated, Pt-Pt10%Rh couples was  $\pm 0.25^\circ$  F.

## FLUIDS

**Irradiated Monoisopropylbiphenyl.** Monoisopropylbiphenyl (MIPB) was irradiated in an in-pile loop at the Reactor and Fuels Research and Development Operations, Hanford Laboratories. The irradiated fluid was heated to 752° F. to stabilize its viscosity. After stabilization, its composition was estimated to be 60% MIPB and 40% tar, where tar is defined as that weight fraction which boils at temperatures above the boiling point of the original fluid.

**Tertiary Eutectic.** This fluid was a mixture of 30.9 mole % biphenyl, 47.5 mole % *o*-terphenyl, and 21.6 mole % *m*-terphenyl with a melting point of 51° F. and a boiling point of 568° F. (4,14).

**Irradiated Tertiary Eutectic.** The tertiary mixture was irradiated at the Hanford facility, and was then heated to 690° F. to stabilize viscosity. After radiation and stabilization of viscosity, the tar content of the fluid was approximately 30%.

***n*-Hexadecane.** This fluid was obtained from Humphrey-Wilkinson as ASTM *n*-cetane.

**Di(2-ethylhexyl) adipate.** This fluid is marketed by Ohio-Apex Division of Food Machinery and Chemical Corporation as Ohio-Apex plasticizer, Adipol 2EH.

**Biphenyl.** Biphenyl was obtained from Monsanto Chemical Co. Melting and boiling points (14) are given as 156° F. and 491° F., respectively.

**MIPB.** This fluid, obtained from Monsanto Chemical Co. was composed of about 62% of the meta isomer and 38% of the para isomer with a melting point of -65° F. and a boiling point of 563° F. (14).

## FILM COEFFICIENTS OF HEAT TRANSFER

**Calculation.** Average values of film coefficients were obtained at the center 12-inch measurement section of the horizontal tubes at steady-state conditions of temperature and fluid velocity with the equation

$$h = \frac{q_{12}}{A_s(t_s - t_b)(1 + \alpha t_s)^2} \quad (1)$$

The heat transferred,  $q_{12}$ , to the fluids from the 12-inch measurement section was measured directly as the wattage

generated in the tube wall, less small radial heat losses of 0.7% to less than 0.2% of the power measured. The heat-transfer area,  $A_s$ , of each tube was measured at room temperature with tube gages and from a volume calibration with mercury. Tubes with uniform inside and outside diameters were selected. Dimensions of the tubes used for each fluid are given in Table I. The mean projection or absolute roughness of the inside surfaces of the tubes was 0.000027 feet as measured from photomicrographs of sectioned tubes.

The average bulk temperature,  $t_b$ , of a fluid was determined as the arithmetical average of the bulk fluid temperatures measured at the entrance and exit mixing pots. The outside tube-wall temperature was measured at nine positions along the length and around the circumference of the measurement section. For any radial position on the circumference, the temperature profile was found to be linear along the tube; however, at any axial position, a temperature difference of 2° to 4° F. from bottom to top was observed around the circumference. This difference was reproducible and was related to fluid velocity, heat flux, and temperature difference ( $t_s - t_b$ ). Earlier work (19) showed that the same average linear temperature profile was obtained with couples located along the top and bottom of a tube, as with couples located along the top, bottom, and each side. In this study, couples were positioned on the top and bottom to give an average linear temperature profile, from which the average outside-wall temperature,  $t_w$ , was determined at the mid-point of each 12-inch measurement section.

Average temperature of the inside tube wall or heat-transfer surface temperature,  $t_s$ , was calculated from Equation 2, in which uniform generation of heat in the tube wall was assumed (2), and properties of Type-347 Stainless steel at average tube wall temperatures were used.

$$q_{12} = \frac{k_w A_s (t_w - t_s)(1 + \alpha t_w)}{0.528 x_w} \quad (2)$$

Calibrations of the flow metering system were made periodically during the course of the experiments; MIPB, the calibrating fluid, was metered between two calibrated tanks. Corrected flowmeter readings were also compared to flows calculated from heat balances across the test section. Flows obtained by these two methods agreed to within  $\pm 3\%$  (20).

**Results.** The film coefficients of heat transfer for the seven fluids are shown in Table II. The loop was operated continuously on a full day basis and each value in the table represents a run composed of a number of readings taken at 2-hour intervals to ensure steady-state conditions and clean-tube values. To prevent boiling during an experiment, loop pressure was maintained from 2 to 3 atm. above a fluid vapor pressure at the highest surface temperature.

## PHYSICAL PROPERTIES

**Density.** Densities were determined with calibrated borosilicate glass dilatometers of the closed type suspended in an air furnace. The dilatometers were calibrated with mercury at room temperature, and appropriate corrections for the expansion of borosilicate glass were made at higher temperatures. The nominal volumes of the dilatometers were 10 and 50 ml. Experimental data for each fluid are presented in Table III.

**Viscosity.** The procedures used for measuring viscosities were fashioned after those described by Miller and others (15). Ostwald-Fenske pipets of the closed type were suspended in a silicone bath at lower temperatures and in an air furnace at higher temperatures. Viscosities were measured for all fluids studied with the exception of *n*-

Table I. Test Section Dimensions<sup>a</sup>

Fluid	Test Section No.	Feet,		Heat Transfer Area, Sq. Ft.
		I.D.,	O.D.,	
MIPB tar mixture	10	0.02628	0.03123	0.08279
Tertiary Eutectic	12	0.02628	0.03120	0.08238
Tertiary Eutectic tar mixture	11	0.02621	0.03115	0.08212
<i>n</i> -Hexadecane	12	0.02628	0.03120	0.08238
Di(2-ethylhexyl) adipate	13	0.02637	0.03132	0.08278
Biphenyl	1	0.02623	0.03167	0.08217
	2	0.02587	0.03164	0.08157
	3	0.02575	0.03162	0.08092
MIPB	3	0.02575	0.03162	0.08092

<sup>a</sup> At room temperature

hexadecane; for this fluid, the values of Rossini (16) were used. Experimental and referenced data for viscosities are given in Table III.

**Specific Heat.** Experimental values of specific heat were used when available. Measurements were made at NRL for irradiated MIPB, MIPB, tertiary eutectic, and biphenyl. The values of specific heat for the irradiated tertiary eutectic were estimated as recommended by Walker and others (21). For *n*-hexadecane and di(2-ethylhexyl) adipate, estimations were based on literature values of similar compounds. The values of specific heat are given in Table III.

**Thermal Conductivities.** Recently Ziebland and Burton (23) measured thermal conductivity values of biphenyl, biphenyl oxide, Dowtherm A, and Santowax R (Santowax R is a proprietary product of the Monsanto Chemical Co. Its composition is 10–15 wt. % *o*-terphenyl, 55–70 wt. % *m*-terphenyl, and 20–30 wt. % *p*-terphenyl) over the approximate temperature range of this work. They included a detailed analysis of error sources and showed that the thermal conductivities of the fluids that were studied decreased about 5 to 10% for each 100° F. increase in temperature. The thermal conductivities of biphenyl and Santowax R were obtained from a smoothed plot of the values of Ziebland and Burton and are given at 100-degree intervals in Table IV. The values for biphenyl were used in correlating the film coefficients of MIPB, and the values for Santowax R were used in correlating the coefficients of the tertiary eutectic and the two irradiated fluids. The thermal conductivity of *n*-hexadecane, which was measured by Sakiadis and Coates (17) at temperatures from 110° to 167° F., and the conductivity of di(2-ethylhexyl) adipate, measured by M.F. Dick (7) at 68° and 140° F., are also included in Table IV. Linear extrapolations were made to the temperature of the NRL experiments for the latter two fluids.

## CORRELATION OF FILM COEFFICIENTS

The film coefficients of heat transfer were correlated by a relation of the Dittus-Boelter type (8), in which the Nusselt number is expressed as a function of the Reynolds number and the Prandtl number with the physical properties of the fluids evaluated at average bulk-fluid temperatures.

$$Nu = A(Re)^a(Pr)^b \quad (3)$$

To facilitate solutions of the constants ( $A$ ,  $a$  and  $b$ ) in this equation, at least three series of experiments were made with each fluid, in which the Prandtl number was maintained constant during any series while the Reynolds number was varied. This was done by maintaining the

average bulk-fluid temperature constant while the fluid velocity and heat-transfer surface temperature were varied. The exponent,  $a$ , of the Reynolds number was obtained directly from the logarithmic plot of the Nusselt number *vs.* the Reynolds number at constant Prandtl number, which, in effect, was a logarithmic plot of film coefficient *vs.* fluid velocity. Since each of the three or more series of experiments for any fluid was made at a different constant Prandtl number, the exponent,  $b$ , of the Prandtl number could then be obtained. The value of the constant,  $A$ , for a fluid was then obtained from a Dittus-Boelter plot of  $Nu$  *vs.*  $(Re)^a (Pr)^b$ .

A best correlation for each fluid was obtained, and these correlations with the range of conditions are given in Table V.

**General Correlation.** The film coefficients measured for the seven fluids were also correlated, Figure 1, by a single

equation, Equation 4, with an average deviation of  $\pm 5.5\%$ .

$$Nu = 0.0175 (Re)^{0.84} (Pr)^{0.4} \quad (4)$$

## FOULING EXPERIMENTS

With polyphenyl fluids in the loop, attempts were made to form scale on the heat-transfer surfaces. These experiments were made at minimum fluid velocity and maximum heat-transfer surface and bulk-fluid temperatures within design limitations of the loop. The maximum heat-transfer surface temperature for a fluid was determined by its vapor pressure. Film coefficient data for the two irradiated fluids indicated a fouling of the heat-transfer surfaces, but the scale was identified as lead and lead oxide by x-ray analyses. This fouling resulted from a lead contamination of the loop which occurred during previous studies with biphenyl polymers that contained lead as an impurity (19). As

(Continued on page 524)

Table II. Film Coefficients of Heat Transfer

Av. Bulk Temp., ° F.	Av. Surface Temp., ° F.	Heat Flux, B.t.u./hr. ft. <sup>2</sup>	Bulk Vel., ft./sec.	Heat-Transfer Coeff., B.t.u./hr. ° F. ft. <sup>2</sup>	$(DV\rho/\mu) \times 10^{-4}$	$c_{p\mu}/k$	$hD/k$	% Dev. Dittus-Boelter, Calcd.-Obsd.
Irradiated Tertiary Eutectic								
624	709	101,400	12.9	1193	6.20	9.17	469.6	-0.2
604	660	64,200	12.9	1147	5.86	9.57	447.1	+2.6
609	690	121,000	17.4	1494	7.94	9.53	584.0	+0.7
610	670	151,500	30.8	2525	14.19	9.46	987.4	-2.8
624	709	101,600	13.0	1195	6.21	9.16	470.4	-0.2
336	482	73,900	8.2	506	1.20	24.87	175.9	+0.5
346	665	165,200	8.2	518	1.29	23.52	181.2	+1.3
342	528	173,200	16.4	931	2.49	24.16	324.6	-0.1
338	453	174,000	29.0	1513	4.35	24.45	526.3	-0.6
520	605	67,800	9.2	798	3.14	12.15	299.9	-1.3
486	651	124,200	9.2	753	2.74	13.50	278.8	-1.3
486	598	144,400	17.0	1289	5.09	13.50	477.0	-2.3
624	706	100,700	13.2	1228	6.33	9.17	483.4	-0.8
628	713	98,800	13.2	1163	6.39	9.11	458.4	+4.6
629	708	99,200	13.5	1257	6.57	9.04	495.7	-1.3
518	587	73,600	13.2	1065	4.41	12.28	399.7	-0.5
520	598	38,500	5.3	494	1.81	12.15	185.5	-0.3
509	831	154,900	5.3	481	1.75	12.48	180.3	-1.4
625	709	100,600	12.7	1198	6.13	9.09	471.9	-2.0
600	744	78,200	5.2	543	2.34	9.63	211.8	-1.4
600	829	365,000	13.0	1233	5.84	9.63	480.7	-5.4
612	853	130,600	5.2	542	2.42	9.40	212.9	0
Tertiary Eutectic								
596	707	248,500	25.0	2251	20.72	5.19	878.2	+3.2
596	629	74,400	25.1	2249	20.80	5.19	876.4	+3.7
585	665	103,200	12.5	1303	10.10	5.29	505.3	+1.2
589	676	245,000	32.1	2806	26.23	5.23	1090.0	+0.9
599	625	83,600	32.2	2835	27.09	5.12	1106.0	-0.5
598	662	252,700	46.1	3918	38.76	5.12	1530.0	-1.6
595	614	73,400	46.0	3861	38.11	5.19	1503.0	-1.3
599	743	85,400	4.6	591	3.87	5.12	230.7	+0.2
452	512	84,700	17.6	1423	8.07	8.37	520.2	+3.1
457	493	84,800	31.1	2322	14.75	8.13	850.7	+1.3
455	476	73,900	48.7	3536	23.89	8.27	1295.0	-0.8
450	614	83,100	4.6	508	2.13	8.35	185.8	-2.1
417	580	222,800	17.5	1366	6.47	10.05	492.6	+2.1
451	557	246,000	30.8	2325	14.16	8.37	850.2	-0.6
446	576	243,800	24.2	1880	10.67	8.43	686.3	-1.7
455	500	84,000	24.0	1887	11.23	8.27	690.7	+0.8
450	563	82,100	7.4	723	3.38	8.42	264.1	+0.6
Irradiated MIPB								
592	689	55,210	5.5	566	2.86	8.46	220.1	+0.4
599	744	82,420	5.7	570	3.04	8.22	222.6	+3.0
602	711	121,800	12.5	1121	6.78	8.17	437.7	+2.4
600	660	122,700	25.0	2038	13.59	8.15	795.8	+0.9
600	686	175,400	25.1	2032	13.63	8.15	793.7	+1.4
602	650	177,000	46.8	3690	25.36	8.17	1440.0	-5.8
587	683	54,970	5.5	574	2.85	8.48	222.8	-0.5
418	524	298,700	45.0	2836	13.24	13.13	1023.0	-3.0
416	582	485,500	45.0	2927	13.13	13.23	1054.0	-6.2
393	542	232,800	23.8	1563	6.41	14.11	557.8	+0.2

Table II. Film Coefficients of Heat Transfer (Continued)

Av. Bulk Temp., ° F.	Av. Surface Temp., ° F.	Heat Flux., B.t.u./hr. ft. <sup>2</sup>	Bulk Vel., ft./sec.	Heat-Transfer Coeff., B.t.u./hr. ° F. ft. <sup>2</sup>	( $DV\rho/\mu$ ) $\times 10^{-4}$	$c_p\mu/k$	$hD/k$	% Dev. Dittus-Boelter, Calcd.-Obsd.
Irradiated MIPB								
408	545	269,200	30.5	1967	8.65	13.57	706.5	-0.2
417	603	223,800	17.0	1203	5.00	13.19	433.8	+1.2
410	505	84,260	12.0	887	3.45	13.45	318.4	+1.8
443	753	289,600	12.2	934	3.91	12.31	341.1	+5.2
592	689	53,740	5.5	557	2.86	8.43	216.8	+1.7
354	457	40,720	5.3	398	1.18	16.42	139.6	+4.0
403	507	43,590	5.1	420	1.43	13.71	150.6	+3.5
401	493	209,650	36.8	2279	10.19	13.82	815.7	+0.1
596	657	184,300	38.6	3028	20.51	8.29	1179.0	-3.0
<i>n</i> -Hexadecane								
217	250	42,000	23.2	1289	4.64	18.55	469.0	+0.3
231	256	42,000	31.4	1679	6.87	17.36	621.0	+1.8
246	266	42,000	39.2	2057	9.30	16.59	778.2	+3.3
259	276	41,900	47.4	2510	11.97	15.95	964.9	+1.3
255	271	42,000	51.7	2709	12.82	16.11	1036	+0.1
243	285	41,400	15.6	996	3.64	16.72	374.7	-0.3
229	298	41,200	8.4	594	1.82	17.52	219.7	-2.4
222	311	41,000	6.0	462	1.25	18.07	169.7	-6.4
286	369	75,100	12.5	907	3.57	14.80	362.5	-2.1
284	416	69,400	6.1	527	1.74	14.90	210.6	-6.3
280	335	71,500	20.1	1300	5.63	14.99	514.4	+0.5
296	341	79,900	28.1	1760	8.46	14.37	711.3	+0.3
303	345	93,000	36.1	2193	11.25	14.09	894.5	+0.1
305	342	97,700	44.0	2599	13.65	14.20	1063	-1.0
300	330	87,800	50.8	2928	15.56	14.26	1191	-1.6
231	248	36,800	42.9	2203	9.29	17.52	814.5	+0.3
398	453	33,300	6.3	609	2.81	11.81	286.9	-4.9
396	446	50,000	12.1	1003	5.37	11.71	466.5	-0.9
392	442	76,400	21.0	1549	9.15	11.95	720.5	+0.3
398	447	98,400	29.1	2025	13.10	11.77	953.6	+0.7
409	456	121,000	37.7	2565	17.48	11.65	1231	-5.4
413	449	108,000	45.2	3003	21.30	11.58	1446	-1.5
402	431	93,800	49.2	3190	22.38	11.75	1513	-1.5
218	251	42,000	23.2	1304	4.68	18.47	475.5	-0.4
Di(2-ethylhexyl) Adipate								
441	461	48,630	36.6	2372	12.35	15.22	1070	-1.2
440	465	48,780	29.2	1967	9.88	15.20	885.4	-1.1
437	469	48,540	22.3	1498	7.60	15.13	673.4	+4.1
434	480	48,360	14.8	1070	4.98	15.27	478.6	+3.1
292	357	124,100	36.0	1909	6.65	23.71	735.2	+2.0
292	370	123,100	28.5	1586	5.27	23.85	614.2	+0.8
309	408	121,700	21.5	1230	4.27	22.59	483.6	+5.0
296	428	117,400	14.5	895	2.72	23.56	347.5	+1.5
281	426	75,400	7.64	518	1.35	24.80	198.2	+0.8
289	368	126,500	28.8	1599	5.27	24.06	617.3	+0.6
442	462	48,180	36.8	2397	12.42	15.27	1085	-2.5
300	428	157,800	21.3	1231	4.08	23.16	479.2	-2.8
411	463	33,170	7.74	640	2.44	15.84	280.2	-1.7
423	496	48,780	7.74	671	2.53	15.47	297.6	-5.5
Biphenyl								
Test Section No. 1								
396	501	100,800	9.4	946	5.62	6.35	360.7	-3.1
421	465	102,800	28.4	2333	18.26	5.98	903.1	+1.7
417	451	102,800	36.9	2940	23.49	6.05	1137	+0.3
416	445	103,100	46.2	3640	29.27	6.05	1405	-2.4
599	685	100,600	9.9	1151	10.35	4.22	508.1	-2.1
585	622	103,200	29.1	2792	29.60	4.31	1219	-0.5
594	665	202,800	29.5	2815	30.62	4.26	1238	+0.3
589	643	202,800	39.2	3690	40.16	4.28	1617	-3.3
585	650	101,700	14.6	1552	14.85	4.31	677.7	+0.3
599	732	202,700	14.7	1514	15.49	4.22	669.1	+4.4
615	747	202,400	14.7	1511	16.04	4.12	676.2	+5.4
608	775	255,100	14.7	1503	15.77	4.16	669.2	+5.3
609	828	342,300	14.7	1544	15.79	4.16	688.9	+2.4
Test Section No. 2								
607	734	203,000	15.5	1581	16.38	4.17	694.1	+4.9
606	781	129,100	5.4	730	5.74	4.17	320.1	-5.8
601	798	145,200	5.7	728	5.93	4.23	318.3	-2.2
605	835	169,300	5.6	724	5.85	4.16	317.1	-3.3
606	831	160,200	5.6	701	5.87	4.17	307.5	+0.2
603	807	142,700	5.6	702	5.92	4.19	307.4	+0.9

Table II. Film Coefficients of Heat Transfer (Continued)

Av. Bulk Temp., ° F.	Av. Surface Temp., ° F.	Heat Flux., B.t.u./hr. ft. <sup>2</sup>	Bulk Vel. ft./sec.	Heat-Transfer Coeff., B.t.u./hr. ° F. ft. <sup>2</sup>	(DV <sub>ρ</sub> /μ) × 10 <sup>-4</sup>	c <sub>p</sub> μ/k	hD/k	% Dev. Dittus-Boelter, Calcd.-Obsd.
Test Section No. 3								
597	659	101,600	15.9	1631	16.26	4.25	702.1	+3.7
602	665	101,600	15.9	1589	16.49	4.21	685.6	+7.2
595	789	147,700	5.7	755	5.82	4.26	328.8	-6.5
600	824	169,900	5.6	750	5.77	4.21	312.7	-2.7
399	571	110,700	5.4	641	3.17	6.30	240.4	-10.3
396	566	107,700	5.4	627	3.15	6.34	233.7	-8.2
500	697	138,600	5.5	693	4.38	5.06	279.3	-7.1
586	739	117,000	5.6	755	5.63	4.31	324.6	-7.3
Monoisopropylbiphenyl								
401	488	100,100	14.3	1139	6.77	8.56	427.9	+1.0
390	445	101,900	26.0	1869	11.79	8.83	696.4	+0.3
398	481	154,700	25.9	1870	12.01	8.68	700.4	+0.6
397	451	128,900	34.2	2359	15.71	8.75	883.1	+0.3
401	424	54,000	34.2	2388	16.45	8.41	896.3	+0.8
407	490	44,800	5.8	542	2.85	8.32	204.2	+0.8
403	587	98,800	5.8	535	2.82	8.35	201.5	+1.4
500	583	103,300	13.9	1237	9.09	6.82	497.9	0.0
507	581	203,000	35.4	2727	23.66	6.71	1103	0.0
499	518	49,800	35.4	2660	23.08	6.82	1069	+1.8
498	603	225,400	26.9	2157	17.57	6.80	866.8	-0.3
505	603	164,900	27.0	2186	17.80	6.78	878.4	-0.6
508	612	64,600	6.1	618	4.06	6.72	250.3	+0.2
600	671	102,000	15.2	1444	13.52	5.51	626.9	+0.3
599	642	102,900	27.5	2392	24.44	5.50	1038	-0.5
603	671	164,100	27.6	2414	25.43	5.32	1051	0.0
599	633	102,500	36.4	3054	32.34	5.51	1325	-1.4
405	487	99,300	14.6	1211	7.12	8.29	456.1	-2.6
398	479	153,900	25.9	1918	12.02	8.69	718.4	-1.8
240	312	102,200	24.9	1418	5.49	15.48	479.5	-0.1
304	530	228,300	14.1	999	4.28	12.07	352.4	-1.9
320	645	151,400	5.7	463	1.85	11.44	165.0	+1.0
600	641	124,100	36.4	3059	33.59	5.30	1328	-0.2
604	678	228,200	36.4	3107	34.98	5.10	1353	-0.4
597	694	110,000	11.5	1129	10.05	5.60	489.1	+0.9
597	816	307,900	14.8	1405	13.21	5.49	609.3	+1.0
599	687	124,400	14.9	1413	13.48	5.40	613.1	+1.2

Table III. Properties of Loop Fluids

Fluids	Temperature, ° F.							Ref.
	100	200	300	400	500	600	700	
Density, Lb./Cu. Ft.								
MIPB Tar Mixture	62.34	59.91	57.49	55.06	52.63	50.20	(47.77)	NRL
Tertiary Eutectic Tar Mixture	67.42	64.92	62.43	59.93	57.43	54.93		NRL
Tertiary Eutectic	65.85	63.08	60.32	57.56	(54.80)	(52.03)		NRL
<i>n</i> -Hexadecane		45.20	42.61	40.06	37.13			(16)
Di(2-ethylhexyl)adipate		53.77	51.17	48.57				NRL
Biphenyl		60.07	57.17	54.27	51.37	(48.47)		NRL(19)
MIPB	61.77	58.93	56.09	53.25	50.41	(47.57)		NRL(19)
Viscosity, Lb./Ft. Hr.								
MIPB Tar Mixture		7.50	3.31	1.90	1.27	0.885		NRL
Tertiary Eutectic		6.06	2.95	1.66	(0.87)	(0.59)		NRL
Tertiary Eutectic Tar Mixture		6.97 <sup>a</sup>	4.94	2.77	1.72	(1.17)	(0.845)	NRL
<i>n</i> -Hexadecane		1.525 <sup>a</sup>	1.320	0.842	0.577			(16)
Di(2-ethylhexyl)adipate		3.43 <sup>a</sup>	2.54	1.52	1.31 <sup>b</sup>			NRL
Biphenyl			1.354	0.857	0.593	(0.438)	(0.340)	(19)
MIPB	11.44	3.21	1.77	1.06	0.73	0.50	(0.33)	(20)
Specific Heat, B.t.u./Lb. ° F.								
MIPB Tar Mixture		0.452	0.496	0.539	0.583	0.627	0.671	(13, 21)
Tertiary Eutectic		0.407	0.453	0.498	0.544	0.489	0.635	(13, 21)
Tertiary Eutectic Tar Mixture			0.466	0.499	0.533	0.566	0.599	(21)
<i>n</i> -Hexadecane		(0.619)	(0.700)	(0.780)	(0.860)	(0.940)		Estd.
Di(2-ethylhexyl)adipate		(0.586)	(0.620)	(0.650)	(0.686)			Estd.
Biphenyl			0.474	0.508	0.542	0.575		(13, 21)
MIPB			0.528	0.566	0.603	0.641		(13, 21)

<sup>a</sup> At 250° F.<sup>b</sup> At 450° F.

Table IV. Thermal Conductivities

Fluids	Temperature, ° F.						Ref.
	200	300	400	500	600	700	
	Thermal Cond., B.t.u./Sq. Ft. Hr. ° F./Ft.						
Biphenyl	0.0782	0.0735	0.0689	0.0643	0.0597	0.0597	(23)
Santowax R		0.0768	0.0738	0.0707	0.0677	0.0597	(23)
<i>n</i> -Hexadecane	0.0729	0.0639	0.0549	0.0459			(17)
Di(2-ethylhexyl)adipate	0.0745	0.068	0.0615	0.055			(7)

Table V. Summary of Correlation Work

Fluid	Best Correlation	Av. Deviation	Range of		Range of Heat Flux, <sup>a</sup>	Range of Av. Fluid Velocity, Ft./sec.
			Reynolds No. × 10 <sup>-4</sup>	Prandtl No.		
Irradiated MIPB	Nu = 0.0140(Re) <sup>0.84</sup> (Pr) <sup>0.49</sup>	± 2.3	1.18-25.4	8.15-16.42	4.07-48.6	5.1-46.8
Irradiated Tertiary Eutectic	Nu = 0.0156(Re) <sup>0.85</sup> (Pr) <sup>0.42</sup>	± 1.4	1.20-14.2	9.04-24.87	3.85-35.6	5.2-30.8
Tertiary Eutectic	Nu = 0.0196(Re) <sup>0.81</sup> (Pr) <sup>0.50</sup>	± 1.5	2.13-38.7	5.12-10.05	7.34-25.3	4.6-48.7
<i>n</i> -Hexadecane	Nu = 0.0292(Re) <sup>0.82</sup> (Pr) <sup>0.3</sup>	± 1.9	1.25-22.4	11.5-18.5	3.33-12.1	6.1-51.7
Di(2-ethylhexyl)-adipate	Nu = 0.0188(Re) <sup>0.84</sup> (Pr) <sup>0.4</sup>	± 2.4	1.35-12.4	15.1-24.8	3.32-15.8	7.6-36.8
Biphenyl	Nu = 0.0174(Re) <sup>0.84</sup> (Pr) <sup>0.39</sup>	± 3.8	3.17-40.2	4.12-6.35	10.1-34.2	5.4-46.2
MIPB	Nu = 0.0138(Re) <sup>0.84</sup> (Pr) <sup>0.47</sup>	± 0.8	1.85-34.9	5.10-15.48	4.48-30.8	5.7-36.4

<sup>a</sup> (B.t.u./hr. sq. ft.) × 10<sup>-4</sup>.

reported in that study, scale could be removed with warm acetic acid or could be swept out at fluid velocities of 20 to 30 feet per second. After a thorough cleaning of the loop and replacement of most of the socket-weld joints, no fouling of any type occurred during experiments with the tertiary eutectic fluid at heat-transfer surface temperatures to 940° F. A summary of the fouling conditions is given in Table VI.

## DISCUSSION

Of the seven fluids studied, the five polyphenyl fluids were of most interest to the program. The two aliphatic fluids were included in an effort to extend the correlation to this type of fluid. To increase the range of fluid properties attempts were made to obtain film coefficients for glycerine and water. However, with glycerine, the bulk-fluid temperature could not be maintained low enough to prevent deterioration of the fluid; and with water, corrosion products from the steel system were deposited on the heat-transfer surface at such a high rate that film coefficients could not be measured.

Film coefficients for the polyphenyl fluids were in the same general range of values for similar conditions of flow and temperature. Coefficients for biphenyl were slightly better than for the fluids of higher molecular weight. However, other considerations such as liquid temperature range, pumping-power requirements, and thermal degradation may be more important in the selection of one of these fluids for a particular application.

A relation of the Dittus-Boelter type, Equation 4, was found to correlate the film coefficients of the fluids more accurately than either a Sieder-Tate (12) or a Colburn (12) type equation. This was demonstrated with each fluid. No significant change in measured film coefficient was observed when film temperatures were varied while bulk-fluid temperature and fluid velocity were maintained constant. With the irradiated tertiary eutectic, the most viscous fluid studied, the temperature potential was increased from 78° to 322° F. at a constant bulk temperature and a constant fluid velocity with no measurable change in the film coefficient. An increase of 11% in the coefficient was predicted by the Sieder-Tate equation, and an increase of 15% was predicted by the Colburn equation.

Table VI. Conditions of Fouling Experiments

Fluid	Surface Temp., ° F.	Bulk-Fluid Temp., ° F.	Fluid Velocity Ft./Sec.	Duration, Hr.
Irradiated MIPB	830	616	5.9	61
	726	602	5.8	70
Irradiated Tertiary Eutectic	850	600	5.5	51
	850	600	5.5	34
Tertiary Eutectic	850	600	5.5	68
	940	620	5.5	76

The reliability of a general correlation is dependent upon the accuracy of the physical property values as well as the film coefficient values and the form of the correlation. The values of density and viscosity are considered acceptable since values for the polyphenyl fluids measured here and by other workers (13, 19) are in agreement. Although specific heat values for each of the polyphenyl fluids were not measured, and exact compositions of the irradiated fluids were not known, the specific heat study of phenyl compounds by Walker (21) made in conjunction with this work, permits an acceptable estimation for these fluids.

In a precise study of the conductivities of four polyphenyl fluids, Ziebland and Burton (23) observed the conductivities to have negative slopes with respect to temperature. This is the same temperature dependency which has been observed by Sakiadis and Coates (17) for a number of organic fluids at lower temperatures, by Briggs (5) for biphenyl to 245° F., and recently by Kerzhentsev and Vargaftik (10) for Dowtherm A to 572° F. The values of conductivity used in correlating the previously published results for biphenyl (19) and (20) were values obtained as part of a preliminary engineering study (13). There was little or no change of conductivity with temperature in these values. The coefficients for biphenyl and MIPB have been recorrelated in this paper using thermal conductivities by Ziebland and Burton. Measured thermal conductivity values for biphenyl were used in correlating the film coefficients of MIPB, since reliable values for MIPB were not available. Conductivity values for the terphenyl mixture, Santowax R, were used in correlating coefficients of the tertiary eutectic and the two irradiating fluids. This

choice was made for the tertiary eutectic since it contained 60 mole % terphenyls. Since the exact compositions of the two irradiated fluids were not known, the effect of the radiation was assumed to result in formation of polyphenyl compounds of higher orders. Thus, the conductivity values of Santowax R were chosen as the most representative of the measured values available.

When this work is compared with the three generally recommended correlations (the Dittus-Boelter, the Sieder-Tate, and the Colburn equations), the limitation of the range of variables in this study is recognized. The importance, however, of accurate property values in the correlation of heat transfer coefficients, and the general lack of such values in the literature, particularly for thermal conductivity, are evident. In fact, McAdams (12) indicated an inadequacy of the form of these correlations, and certainly some degree of this inadequacy must be due to uncertainty of property values.

In arriving at a general correlation, more weight was given to the biphenyl data since the physical properties used in the correlation for this fluid had been measured for the temperature range involved. Also, the heat-transfer data obtained for biphenyl by Silberberg and Huber (18) and by NRL agree, provided the same property values are used in the comparison. Further, this equation is almost identical to an equation obtained by Kaufman and Isley (9) in a study of heat transferred to water at Reynolds numbers between 10,000 and 50,000.

#### ACKNOWLEDGMENT

The authors are grateful for appreciable help in the continuous operation and necessary alterations in the loop equipment. They are particularly indebted to the following, who contributed much to the successful operation, measurements, and calculations: C.H. Blachly, J.R. Spann, E.W. Steinkuller and B.E. Walker, and Mrs. J.B. Burbank of the Electrochemistry Branch for the x-ray analysis and identification of the lead oxide scale.

#### NOMENCLATURE

$A$  = area, sq. ft.  
 $A_1$  = log mean area of test-section wall  
 $A_2$  = area of heat-transfer surface  
 $c_p$  = specific heat, B.t.u./lb., ° F.  
 $D$  = diameter, feet  
 $h$  = film coefficient of heat transfer, B.t.u./hr. sq. ft., ° F.  
 $k$  = thermal conductivity, B.t.u./hr. ft., ° F.  
 $k_w$  = thermal conductivity of test-section wall at average wall temperature  
 $q_{12}$  = heat input to the 12-inch section of tube, B.t.u. per hour  
 $t$  = temperature, ° F.  
 $t_b$  = average bulk temperature of fluid

$t_w$  = average outside test-section wall temperature  
 $t_s$  = average heat transfer surface temperature  
 $V$  = fluid velocity, ft./hr.  
 $x_w$  = wall thickness of test section, feet  
 $\alpha$  = average coefficient of thermal expansion (Type 347 Stainless steel) from room temperature  
 $\rho$  = density, lb./cu. ft.  
 $\mu$  = absolute viscosity, lb./ft. hr.

#### LITERATURE CITED

- (1) Anderson, K., Argonne Natl. Lab. Rept. 5121 (August 1953).
- (2) Bernardo, E., Eian, C.S., *Natl. Advisory Comm. Aeronaut.*, Rept. ARR No. E5F07, E-136 (August 1945).
- (3) Bolt, R.O., Carrol, J.G., Fontana, B.J., U.S.A.E.C., Rept. TID-5148, October 1953.
- (4) Bowen, H.C., Groot, C., Hanford Atomic Products Operation, Rept. HW-48427, Feb. 13, 1957.
- (5) Briggs, D.K.H., *Ind. Eng. Chem.* 49, 419 (1957).
- (6) DeHalas, D.R., Hanford Atomic Products Operations, Rept. HW-53718, November 1957.
- (7) Dick, M.F., Univ. Microfilms, Pub. No. 3488; Dissertation Absts., (Univ. of Michigan, Ann Arbor) 12, 166-7 (1952).
- (8) Dittus, F.W., Boelter, L.M.K., *Univ. Calif. Pub. Eng.* 2, 443 (1930).
- (9) Kaufman, S.J., Iseley, F.D., *Natl. Advisory Comm. Aeronaut.*, Research Memo RM E50G31 Sept. 27, 1950.
- (10) Kertzhentsev, V.V., Vargaftik, N.B., *Ihimicheskaya Promyshlennost*, Part 3, 82-4 (1950) (AERE Lib/Trans 809).
- (11) Loftness, R.L., North American Aviation Rept. NAA-SR-280, December 1953.
- (12) McAdams, W.H., "Heat Transmission," 3rd. ed., p. 219, McGraw-Hill, New York, 1954.
- (13) McEwen, M., "Preliminary Engineering Study of Organic Nuclear Reactor Coolant-Moderators," Monsanto Chemical Co., March 31, 1956.
- (14) McEwen, M., "Organic Coolant Databook," *Ibid.*, St. Louis 24, Mo., 1958.
- (15) Miller, R.R., Ewing, C.T., Hartman, R.S., Atkinson, H.B., Jr., Naval Research Lab. Rept. C-3105, 3-4 (April 1947).
- (16) Rossini, F.D., Ptizer, K.S., Arnett, R.L., Pimentel, G.C., Braum, R.M., "Selected Values of Physical and Thermodynamic Properties of Hydrocarbons and Related Compounds," pp. 228-9, Carnegie Press, Pittsburgh, Pa., 1953.
- (17) Sakiadis, B.C., Coates, J., *A.I.Ch.E. Journal* 3, 121-22 (1957).
- (18) Silberberg, M., Huber, D.A., *Atomics Internatnl Rept. NAA-SR-2796*, January 1959.
- (19) Stone, J.P., Ewing, C.T., Blachly, C.H., Walker, B.E., Miller, R.R., *Ind. Eng. Chem.* 50, 895-902 (1958).
- (20) Stone, J.P., Ewing, C.T., Blachly, C.H., Steinkuller, E.W., Miller, R.R., Naval Research Lab. Rept. 5225, November 1958.
- (21) Walker, B.E., Jr., Brooks, M.S., Ewing, C.T., Miller, R.R., *J. CHEM. ENG. DATA* 3, 280-2 (1958).
- (22) Wheelock, C.W., *Atomics Internatnl. Rept. NAA-SR-2558*, August 1957.
- (23) Ziebland, H., Burton, J.T.A., *J. CHEM. ENG. DATA* 6, 579-83 (1961).

RECEIVED for review January 23, 1962. Accepted June 15, 1962.