GENERALIZATION OF EXPERIMENTAL RESULTS ON THE THERMAL CONDUCTIVITY OF FREONS 21, 22, AND 23

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A system of computational equations is developed and handbook tables for the thermal conductivity of Freons 21, 22 and 23 are compiled in a broad range of state parameters.

Existing handbook data on the thermal conductivity of Freons 21, 22, and 23 [1-7] are based on limited information and afford a possibility of determining λ for gaseous Freons only at atmospheric pressure and for liquid Freons near the saturation curve. Meanwhile, quite extensive experimental material on the thermal conductivity of the mentioned Freons has been accumulated up to now in a broad range of temperatures and pressures, including the rarefied and dense gas domain and for a liquid down to the crystallization curve. Hence, the purpose of this paper is to analyze existing experimental data on the thermal conductivity of Freons 21, 22, and 23, to extract the most reliable results, to develop a system of computational equations, and to compile tables of handbook data in a broad range of state parameters on this basis.

The greatest number of experimental researches on the thermal conductivity of the Freon group under consideration refer to Freon 22. A list of these paper is presented in Table 1.

To describe the thermal conductivity λ_t of gaseous Freon 22 at atmospheric pressure, we took account of the results of investigations [8, 15, 21, 23], which agreed within 3-5% limits and are assumed equally exact. The initial array of $\{\lambda_t, T\}$ data was composed of 37 experimental points in the 251-450°K temperature range for the joint processing of these results. The equation for λ_t is represented in the domain under consideration for the parameters in the form

$$\lambda_{\text{т}} = \sum_{i=0}^{1} a_i T^i. \tag{1}$$

The comparison between experimental values of λ_t and values computed by means of (1) is presented in Fig. 1.

There are only data [23] at pressures $p > p_s$ in the dense gas and liquid domains, which we processed in the form of a dependence of the excess thermal conductivity on the density:

$$\lambda - \lambda_{\text{т}} = \sum_{i=1}^{4} b_i \rho^i; \tag{2}$$

hence, the array of $\{\lambda, \rho\}$ data included 118 experimental values of the thermal conductivity in the 10-900-kg/m³-density range.

The dependence of the excess thermal conductivity of Freon 22 on the density is unique in this range of parameters (for $\rho \leq 900$ kg/m³). At higher densities stratification of the excess thermal-conductivity isotherms occurs in the Δλ, ρ-coordinates and hence the following scheme was selected to generalize the data of liquid Freon 22 with respect to λ. Taking account of the specifics of using Freons as working bodies of refrigerators, for which the

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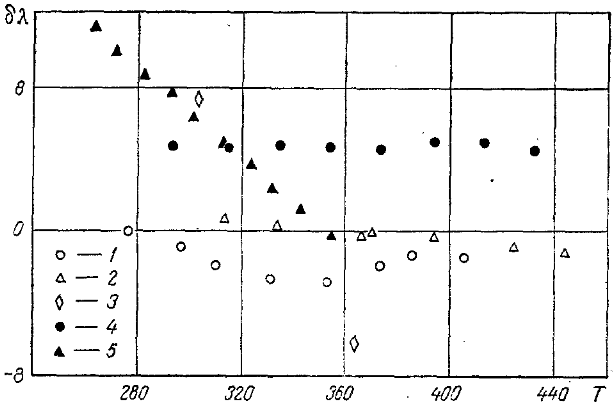

Fig. 1. Deviation (in %) of the experimental values of the thermal conductivity of gaseous Freon 22 at atmospheric pressure from the values computed by means of (1) as a function of the temperature (°K). 1) Data in [15]; 2) [21]; 3) [10]; 4) [23]; 5) [8].

data on the thermophysical properties are especially important near the elasticity curve, experimental values of the thermal conductivity along the saturation line were processed primarily in compiling the system of computational equations for λ, and the pressure dependence of the thermal conductivity was determined by means of results from [23, 24] (the pressure dependence of the thermal conductivity of liquid Freon 22 was not studied in other papers).

It should be noted that existing methods of generalizing experimental results based on the selection of appropriate weights for each group according to the degree of their confidence can apparently result in affirmative results in the presence of initial quantities in satisfactory agreement and a relatively uniform distribution of the experimental points in the range of state parameters under consideration according to the data of different authors. Meanwhile, an analysis of the data on the thermal conductivity of liquid Freon 22 presented in Table 1 near the saturation line shows that it is impossible to consider the agreement between the mentioned results satisfactory in either their absolute values or, even more, their temperature history. The concentration of the experimental results turns out to be highest in the 230-300°K range, where the results in [14, 16] (which are lower compared to other data) have the greatest number of test points in this domain. Therefore, taking the average of the data of different authors even with their weights taken into account more or less objectively can result in substantial distortion of the temperature dependence of λ and significant errors during extrapolation toward low and high temperatures.

In connection with the above, we selected the following scheme in the computation of λ values to be recommended for Freon 22 in the boiling liquid state. The temperature dependence of the thermal conductivity was taken on the basis of two independent series of experiments performed in the M. V. Lomonosov Odessa Technological Institute [23, 24]. These measurements were made by an absolute stationary hot-wire method taking account of all its inherent corrections, include the broadest temperature range (from 113-353°K), and agree well enough with the most reliable results of other authors.

All the experimental material was divided into three groups to match the dependence obtained with the results of other measurements and to clarify the most probable values of λ_s. In the first group were results whose error was 1-2%, while in the second group it was 2-3%, and in the third group 3-5% (unfortunately, it is not possible to present a detailed analysis of the experimental research on whose basis the error in the data was estimated because of limited space in the paper). Results from [9], with the exception of the 310-343°K range (where an anomalous temperature history of the thermal conductivity is observed), as well as data from [22] were in the first group, while results from [8, 12, 13, 17, 20] comprised the second group, and [14] the third group. Results from [10, 11, 16] were excluded from consideration, since they differ substantially from all the remaining measurements.

The deviations of the experimental results of the papers mentioned from the dependence $\lambda_s - T$ obtained in processing the data in [23, 24] were then computed. To take the average

TABLE 1. Papers Investigating the Thermal Conductivity of Freon 22

Reference	Phase	Temp. range, °K	Pressure range, MPa	Method	No. of test points
[10]	G	303—363	0,1	w	2
	L	273—313	$p \sim p_s$	c	2
[8]	G	251—351	0,16—1,06	w	48
	L	198—291	$p \sim p_s$	w	68
[11]	L	253—293	$p \sim p_s$	p	3
[12]	L	193—296	$p \sim p_s$	nw	6
[13]	L	153—208	$p \sim p_s$	nw	6
[14]	L	236—304	$p \sim p_s$	nw	8
[15]	G	278—407	0,003—0,12	w	8
[16]	L	248—287	$p \sim p_s$	nw	4
[17]	L	216—271	$p \sim p_s$	w	4
[9]	L	193—343	$p \sim p_s$	r	—
[18]	L	256	$p \sim p_s$	com	1
[19]	L	216—271	$p \sim p_s$	w	4
[20]	L	148—303	$p \sim p_s$	nw	13
[21]	G	315—445	0,15—1,6	c	16
[22]	L	126—227	$p \sim p_s$	w	14
[23]	G, L	193—433	0,1—59	w	158
[24]	L	113—295	0,1—59	w	91

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[*] Provisional notation: c) coaxial cylinder method; w) hotwire method; p) plane-layer method; nw) nonstationary hot-wire method; r) regular thermal mode method; com) thermal comparator method.

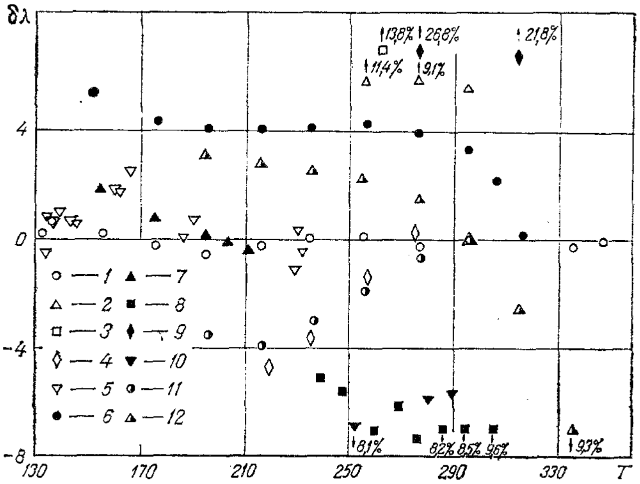

Fig. 2. Deviation (in %) of experimental values of the thermal conductivity of liquid Freon 22 along the saturation line from those computed by means of (3) as a function of the temperature (°K): 1) data from [23, 24]; 2) [11]; 3) [18]; 4 [17]; 5 [22]; 6) [20]; 7) [13]; 8) [14]; 9) [10]; 10) [16]; 11) [8]; 12) [9].

of the deviations obtained, these latter were added with the signs taken into account, where the deviations of results from the first group of experimental data were taken into account with weight 1, the second with weight 0.5, and the third with weight 0.25. The absolute values of λ_s were then corrected by the magnitude of the mean deviation found in this way and hence the temperature dependence of the thermal conductivity (i.e., the quantity $[\partial\lambda/\partial T]_p$ determined from the results of [23, 24]) was conserved.

The initial array of $\{\lambda_s, T\}$ data compiled for the statistical processing by using an electronic computer included 113 experimental values of λ_s in the 113-360°K range. The equa-

TABLE 2. Coefficients of (4)

j	i			
	0	1	2	3
0	—1,19704	—0,68156	1,40965	—0,94426
1	—0,33919	2,25599	—5,05850	4,45291
2	—3,67110	21,18587	—32,63514	16,91989
3	9,77897	—56,31672	83,18183	—49,13515
4	—10,37926	59,36453	—79,97439	51,61741
5	5,12614	—29,14068	35,19539	—24,96465
6	—0,97589	5,52094	—5,29561	4,66327

TABLE 3. Coefficients of (1), (2), and (3)

Coefficients	Freon 21	Freon 22	Freon 23
a_0	—6,378·10^{-3}	—8,269·10^{-3}	—2,880·10^{-3}
a_1	5,135·10^{-5}	6,330·10^{-5}	5,425·10^{-5}
b_1	0,026504	0,0340072	0,051855
b_2	0,038283	—0,000541	—0,012754
b_3	—0,041032	0,028852	0,047425
b_4	0,026114	—0,007593	—0,021362
c_1	—0,22593	0,53089	0,04288
c_2	2,87771	—1,06620	1,86268
c_3	—5,26325	4,58756	—2,08494
c_4	4,03891	—8,36907	—1.29820
c_5	—0,98069	6,92396	4.16977
c_6	—0;00627	—1,98931	—1,82558

tion for λ_s obtained on the basis of the principles mentioned is represented in the form*

$$\lambda_s = \lambda_{cr} + \sum_{i=1}^{6} c_i (T_{cr} - T)^{i/3}. \tag{3}$$

For $T = T_{cr}$, $\lambda = \lambda_{cr}$, found from (2) and satisfying the condition $(\partial\lambda/\partial T)_{cr} \to \infty$. Deviations of the experimental results of different authors from the λ_s values of Freon 22 computed by means of (3) are presented in Fig. 2.

The pressure dependence of the thermal conductivity was determined by the following equation generalized for the Freon group under consideration:

$$(\lambda/\lambda_s)_T = \sum_{i=0}^{3}\sum_{j=0}^{6} a_{ij} (T/T_{cr})^i (p/10p_{cr})^j, \tag{4}$$

for whose compilation more than 270 $\{\lambda, p, T\}$ values were used (including 125 for Freon 22) in the ranges $\tau = 0.3-1$ and $\pi = 1-12$. The coefficients of (4) are presented inTable 2.

Values of the thermal-conductivity coefficient computed by means of (4) agree with the experimental results in [23, 24] with an error not greater than the error in the test. It should be noted that (3) and (4) are valid for the whole liquid state domain down to the crystallization curve and pressures to 50-60 MPa.

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*The author knows of information on the anomalous behavior of the thermal conductivity in the critical domain which is expressed by the appearance of thermal conductivity maxima in the $\Delta\lambda$, ρ coordinates. Therefore, the value of λ_{cr} found by means of (2) does not correspond to the true magnitude of the thermal conductivity at the critical point. However, it should be noted that (3) can be transformed in an appropriate manner after accumulation of experimental results in this domain. The range of operation of (3) should be limited to the temperature 360-365°K.

TABLE 4. Thermal Conductivity $\lambda \cdot 10^4$ of Freon 21 [W/(m·°K)]

Temperature, °C	Pressure, MPa									
	0,1	1	2	4	6	8	10	12	16	20
0	1090	1094	1098	1108	1118	1127	1135	1142	1152	1158
10	81,8	1057	1061	1071	1082	1092	1100	1108	1118	1124
20	86,9	1020	1024	1035	1046	1056	1066	1073	1084	1090
30	92,0	983	988	999	1011	1022	1031	1039	1050	1056
40	97,2	946	952	964	976	988	998	1006	1018	1024
50	102,3	910	916	929	942	954	965	974	986	993
60	107,5	874	881	895	909	921	933	942	955	963
70	112,6	839	846	861	876	889	901	911	926	934
80	117,7	804	812	828	844	858	871	882	897	906
90	122,9	134,2	778	796	312	828	842	853	870	880
100	128,0	138,8	746	764	782	799	813	826	844	856
110	133,2	143,5	714	734	753	771	786	800	820	833
120	138,3	148,2	683	704	724	743	760	774	796	811
130	143,4	153,0	167,1	674	696	716	734	750	774	790
140	148,6	157,8	170,9	645	668	689	709	725	752	769
150	153,7	162,6	174,8	613	638	660	681	699	727	749
160	158,9	167,5	178,9	561	605	633	655	672	704	730
170	164,0	172,3	183,2	224	562	600	627	645	681	709
180	169,1	177,2	187,5	222	501	564	598	618	659	689
190	174,3	182,1	191,9	223	346	520	566	590	637	670
200	179,4	187,1	196,4	224	290	465	532	560	615	651

TABLE 5. Thermal Conductivity $\lambda \cdot 10^4$ of Freon 22 [W/(m·°K)]

Temperature, °C	Pressure, MPa									
	0,1	1	2	4	6	8	10	12	16	20
—40	66,5	1156	1162	1174	1186	1197	1206	1214	1226	1232
—30	72,7	1108	1114	1126	1139	1151	1161	1170	1181	1188
—20	79,0	1059	1065	1079	1093	1106	1116	1125	1137	1144
—10	85,3	1010	1017	1032	1047	1061	1072	1082	1095	1103
0	91,5	962	970	986	1002	1017	1029	1040	1054	1062
10	97,8	915	923	941	958	974	987	999	1014	1023
20	104,1	868	877	896	915	932	947	959	977	987
30	110,4	123.1	832	853	873	892	908	921	940	953
40	116,7	128,6	788	810	832	852	870	884	906	920
50	123,0	134,3	744	768	792	814	832	849	873	889
60	129,3	140,1	156,4	726	751	774	795	812	839	858
70	135,6	145,9	160,8	680	707	732	753	773	803	823
80	141,9	151,8	165,6	610	654	683	706	726	759	787
90	148,2	157,8	170,6	217	596	639	669	694	730	761
100	154,5	163,7	175,8	214	502	587	628	658	701	735
110	160,8	169,7	181,2	214	316	521	584	621	672	710
120	167,1	175,7	186,7	216	275	433	532	582	643	685
130	173,4	177,0	192,3	219	264	361	474	540	614	660
140	179,7	187,8	197,9	223	260	326	420	497	584	636
150	186,0	193,9	204	227	259	309	382	456	554	612
160	192,3	200	209	231	260	302	359	423	525	589
170	198,6	206	215	236	263	298	341	399	498	566
180	205	212	221	241	266	297	336	383	474	545
190	211	218	227	246	269	297	332	372	455	525
200	218	224	233	251	273	299	330	365	440	507

Systems of computational equations of λ for the other Freons of this group were compiled by an analogous method. The initial array of $\{\lambda_t, T\}$ data for Freon 21 included 31 experimental values of λ_t in the 240-465°K range according to the results of [10, 15, 29], the array of $\{\lambda, \rho\}$ data included 36 values of λ in the 10-230 kg/m^3 density range according to results in [29], and the array of $\{\lambda_s, T\}$ data included 40 values of λ_s in the 148-432°K temperature range according to the results in [17, 20, 29, 30]. A total of 50 $\{\lambda, p, T\}$ values in the 210-432°K temperature and 5-59-MPa-pressure ranges according to the data in [30] were used in compiling (4).

The information about the $\{\lambda_t, T\}$ and $\{\lambda, \rho\}$ dependences for Freon 23 was borrowed from [31] (12 and 94 experimental values, respectively, in the 283-435°K temperature and 10-800 kg/m^3 density ranges). Data in [20] in the 148-268°K range (8 experimental points) and in [24, 31] in the 118-292°K range (32 experimental points) were used to describe the thermal conductivity of liquid Freon 23 along the saturation curve. Discrepancies between these re-

TABLE 6. Thermal Conductivity $\lambda \cdot 10^4$ of Freon 23 [W/(m·°K)]

Temperature, °C	Pressure, MPa									
	0,1	1	2	4	6	8	10	12	16	20
—80	75,9	1338	1345	1360	1375	1389	1401	1411	1423	1430
—70	81,3	1262	1270	1287	1303	1318	1331	1341	1355	1362
—60	86,7	1186	1195	1213	1231	1247	1261	1272	1287	1295
—50	92,1	1110	1119	1139	1159	1176	1191	1204	1220	1229
—40	97,6	1033	1044	1066	1087	1106	1123	1136	1155	1166
—30	103,0	124,8	969	993	1017	1038	1056	1071	1092	1105
—20	108,4	128,6	896	922	948	970	990	1007	1031	1046
—10	113,8	132,8	824	852	880	905	926	945	972	990
0	119,2	137,1	161,5	782	811	838	862	882	913	934
10	124,7	141,6	163,4	706	737	766	791	813	847	870
20	130,1	146,3	166,1	245	715	757	786	809	844	871
30	135,5	151,0	169,3	227	593	686	731	763	807	839
40	140,9	155,8	172,9	221	346	588	665	710	767	806
50	146,3	160,6	176,7	219	293	460	586	650	724	771
60	151,8	165,5	180,8	219	276	377	503	586	679	735
70	157,2	170,5	185,0	220	268	339	437	525	634	698
80	162,6	175,4	189,3	222	263	320	394	473	590	662
90	168,0	180,4	193,8	224	261	309	369	435	549	627
100	173,4	185,5	198,3	227	261	302	352	408	515	595
110	178,9	190,6	203	230	261	298	342	390	487	565
120	184,3	195,6	208	233	263	296	335	377	464	540
130	189,7	201	212	237	265	296	330	368	447	519
140	195,1	206	217	241	267	296	327	361	433	500
150	200	211	222	245	270	297	326	357	422	486
160	206	216	227	249	272	298	325	354	414	473
170	211	221	232	253	276	300	325	352	408	463
180	217	227	236	257	279	302	326	351	404	455
190	222	232	241	261	282	304	327	351	399	448
200	228	237	246	266	286	306	328	350	395	440

TABLE 7. Thermal Conductivity $\lambda \cdot 10^4$ of Freons 21, 22, and 23 during the Saturation Time [W/(m·°K)]

Temper-ature, °C	Freon 21		Freon 22		Freon 23		Tempre-ature, °C	Freon 21		Freon 22		Freon 23	
	λ'	λ''	λ'	λ''	λ'	λ''		λ'	λ''	λ'	λ''	λ'	λ''
—80	—	—	—	—	1331	78,7	50	905	106,6	742	151,3	—	—
—70	—	—	—	—	1257	85,9	60	869	113,2	703	166,6	—	—
—60	—	—	—	—	1183	93,9	70	836	120,0	658	185,2	—	—
—50	—	—	—	—	1109	102,9	80	804	127,3	609	210	—	—
—40	—	—	1150	66,9	1032	113,1	90	770	135,1	533	250	—	—
—30	—	—	1102	73,8	958	125,2	100	741	143,5	—	—	—	—
—20	—	—	1055	81,2	891	139,5	110	712	152,6	—	—	—	—
—10	—	—	1008	89,1	822	157,1	120	681	162,8	—	—	—	—
0	1087	77,4	956	97,5	753	179,7	130	654	174,4	—	—	—	—
10	1050	82,9	912	106,5	683	211	140	625	187,8	—	—	—	—
20	1013	88,6	866	116,1	587	264	150	596	204	—	—	—	—
30	976	94,4	824	126,6	—	—	160	562	225	—	—	—	—
40	941	100,4	784	138,2	—	—	170	508	257	—	—	—	—

sults do not exceed 2.5% and hence were assumed equally exact during the processing. A total of 96 test {λ, p, T} values of Freon 23 in the 118-290°K temperature and the 59 MPa pressure p_s ranges according to data in [24, 31] were used in forming the generalized equation (4).

The coefficients of (1), (2), and (3) are represented in Table 3, while values of thermal conductivity of Freons 21, 22, and 23 in a single-phase domain are presented in Tables 4-6. These tables are computed in the temperature range from the normal boiling point to 200°C at pressures up to 20 MPa.* Values of the thermal conductivity along the saturation curve (according to the temperatures) are given in Table 7.

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*The system of equations proposed can be used to compute the thermal-conductivity equations down to the crystallization curve at pressures to 50-60 MPa.

It should be noted that investigations performed in recent years for a number of substances show that the uniqueness of the dependence $\Delta\lambda - \rho$ is not conserved near the saturation curve on the vapor side, in which connection the quantities λ'' presented in Table 7, just as the values of the thermal conductivity in the critical domain, should be considered approximate. The error in the recommended values of the thermal conductivity of Freons in the rest of the parameter domain apparently does not exceed 3-5%.

NOTATION

λ, coefficient of thermal conductivity; λ_t, coefficient of thermal conductivity in the gas phase at atmospheric pressure; λ_s, λ', and λ'', coefficients of thermal conductivity on the saturation line; λ_{cr}, critical coefficient of thermal conductivity; T, temperature; p_s, saturation pressure; T_{cr}, critical temperature; ρ, density; a, b, c, α, coefficients.

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EXPERIMENTAL INVESTIGATION OF THE THERMAL CONDUCTIVITY OF A BINARY Ar—Kr MIXTURE AT LOW TEMPERATURES

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New experimental results on the thermal conductivity of an Ar—Kr mixture in the 120-273°K temperature range are obtained.

There is a limited quantity of experimental results on the thermal conductivity of binary mixtures of monatomic gases in the $T \leq 273$°K temperature range at atmospheric pressure. Wachsmuth [1] measured the thermal conductivity of an He—Ar mixture at atmospheric pressure and a temperature near 273°K (273.1°K), as did Rychkova and Golubev [2] within the limits 196.66-372.86°K and pressures from 0.1-29.4 $\mathrm{mN/m^2}$. The authors of [2] obtained values of the thermal conductivity for four compositions of a helium—argon mixture in the low-temperature measurement range at 196.66°K and at pressures from 0.1-29.4 $\mathrm{mN/m^2}$. Davidson and Music [3] measured the thermal conductivity of a helium—neon mixture at 273.1°K at atmospheric pressure, and Srivastava and Madan [4] performed the measurements for a neon—argon mixture.

We first investigated the thermal conductivity of an Ar—Kr mixture experimentally in the $T = 120$-273°K temperature range at atmospheric pressure for five argon concentrations: 25, 50, 75, 90.15, and 98.5%. High-purity argon with a 99.987% content of the main substance and 99.97% krypton were used in the tests. The measurements were performed on an apparatus employing the absolute hot-wire method [5].

Experimental results of the temperature dependences of the thermal conductivity of the mixture on composition are presented in Table 1; the following notation is used: Q_H is the quantity of heat transferred by conduction from the wire through the layer of mixture under investigation to the inner wall of the measuring tube; T_1 and T_2 are readings of the inner and outer resistance thermometers; Q is the total quantity of heat liberated by the heater; Q_R is the quantity of heat transferred by radiation from the heater to the wall of the measuring tube; Q_C is the quantity of heat transferred along the current supplying and potential conductors; λ is the thermal conductivity of the mixture; T_{av} is the reference temperature; and ΔT_{mix} is the temperature drop in the mixture layer under investigation.

Corrections for heat removal from the ends of the heater, radiation, and the temperature drop in the wall of the glass tube of the measuring cell were taken into account in processing the measurement results.

As measurements and calculations showed, corrections to the readings of the inner thermometer according to the outer for nonheating currents before each measurement (ΔT_{grad}), the temperature jump, eccentricity, and change in geometric size of the cell with temperature lie within the limits of experimental accuracy in the calculation of the thermal conductivity and hence were not introduced.

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