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THERMAL CONDUCTIVITY OF KETONES AS A FUNCTION OF TEMPERATURE AND PRESSURE

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UDC 536.22+536.23

Results are presented from an experimental study of thermal conductivity of ketones in the liquid and gaseous states at various temperatures and pressures, including the range of critical and supercritical state parameters.

Analysis of published studies of ketone thermal conductivity shows that this property has been studied mainly as a function of temperature at atmospheric pressure [1, 2]. The results of various studies [2-5] of acetone thermal conductivity versus temperature diverge both qualitatively and quantitatively. At 293°K this divergence is 4%, while at 313°K it reaches 12%.

In the present study measurements were made of the thermal conductivity of liquid and gaseous dimethylketone (acetone), methylethylketone, methylbutylketone, and dipropylketone over the temperature range of 285.9-713.3°K at pressures of $(0.98\text{-}490)\cdot 10^5$ Pa. The measurements were performed by the cylindrical calorimeter regular thermal regime method [6, 7]. In the liquid phase the thickness of the layer studied was 0.625 and 0.55 mm, while in the gas phase thicknesses of 0.36 and 0.43 mm were used. The temperature head at the specimen boundary varied from 1.7 to 0.5°K.

Thermal conductivity measurements performed with differing temperature differentials at the specimen boundaries revealed that there was no convective heat exchange in the experiments, as evidenced by the good agreement of all the experimental data. For all measurements the product of the Grashof and Prandtl numbers Gr•Pr was less than 1000, which also confirmed the absence of any convective effect on the thermal conductivity values.

Comparison of the present experimental data on thermal conductivity of liquid ketones at atmospheric pressure with the recommended values of [1] revealed that they coincide to an accuracy of 1-2%. The maximum relative uncertainty of the measurements did not exceed 2.5%, except for the critical region where it increased somewhat to 3-4.5%.

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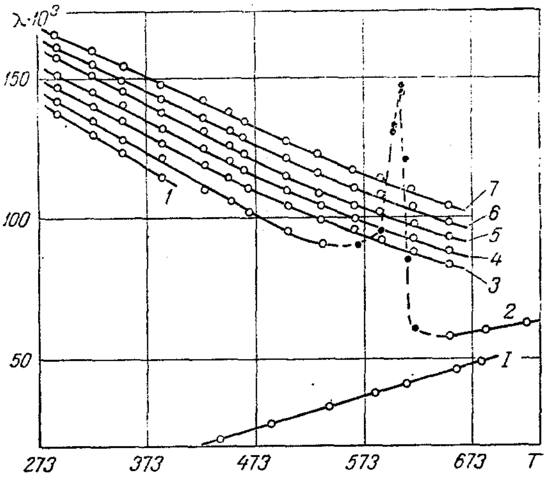

Fig. 1. Thermal conductivity of dipropylketone versus temperature at P•(10^5 Pa): 1) 0.98; 2) 28.2; 3) 98; 4) 196; 5) 294; 6) 392; 7) 490. $\lambda \cdot 10^3$, W/m•deg; T, °K.

Table 1 shows experimental data for thermal conductivity of liquid ketones. Figure 1 depicts the change in thermal conductivity of dipropylketone as a function of temperature and pressure.

The thermal conductivity coefficient of liquid ketones decreases with increase in temperature over the entire pressure range studied, with the exception of the critical region, while with increase in pressure the absolute value of the coefficient increases.

To evaluate the effect of temperature on thermal conductivity, Table 2 presents values of $(\partial\lambda/\partial T)_P$ for liquid ketones.

With increase in excess pressure and increase in temperature there is a decrease in the derivative $(\partial\lambda/\partial T)_P$. With increase in number of carbon atoms for the lower members of the homological series $(\partial\lambda/\partial T)_P$ decreases, while with further increase in the length of the carbon chain, there is some increase in the derivative.

To evaluate the effect of excess pressure on liquid ketone thermal conductivity Table 2 presents values of the derivative $(\partial\lambda/\partial P)_T$. With increase in temperature and decrease in pressure the value of $(\partial\lambda/\partial P)_T$ increases in ketones.

A similar pattern of change in $(\partial\lambda/\partial T)_P$ and $(\partial\lambda/\partial P)_T$ has been observed in a study of thermal conductivity of saturated hydrocarbons and saturated monatomic alcohols [8, 9].

With increase in molecular mass the thermal conductivity of liquid ketones decreases.

For measurements in the critical region along the isobars $49 \cdot 10^5$ Pa for acetone, $39.2 \cdot 10^5$ Pa for methylethylketone, $31.5 \cdot 10^5$ Pa for methylbutylketone and $28.2 \cdot 10^5$ Pa for dipropylketone there was an anomolous increase in the thermal conductivity coefficient, shown by the dashed line of Fig. 1.

In the critical region measurements were performed in steps of 0.1-5°K and various temperature differentials across the layer, for example, 1.7-1 and 1.01-0.5°K for acetone. The experimental data obtained for the various temperatures varied over 3-4%.

Thermal conductivity of gaseous ketones was studied over the temperature range 340.5-713.3°K at atmospheric pressure. With increase in temperature the thermal conductivity coefficient of all the gaseous ketones increased linearly (Fig. 1, line 1).

According to [1], only the thermal conductivity of gaseous dimethylketone has been studied previously, over a narrow temperature interval. The present data agree with acetone vapor values recommended in [1] within 1-2%.

To generalize the experimental thermal conductivity, data for liquid and gaseous ketones the law of corresponding states was used in the following form:

$$\lambda_T/\lambda_1 = f(T/T_{cr}), \tag{1}$$

TABLE 1. Thermal Conductivity of Liquid Ketones at Various Temperatures and Pressures ($\lambda\cdot 10^3$, W/m·deg)

T, K	λ
Acetone	
$P=0{,}98\cdot 10^5$ Pa	
288,7	164,2
311,2	154,3
$P=49\cdot 10^5$ Pa	
288,7	166,7
311,2	158,1
338,1	146,9
359,0	135,2
391,0	123,9
423,0	111,2
463,9	96,6
488,5	89,4
494,3	89,8
498,3	90,3
502,4	92,1
504,0	99,7
506,8	120,2
508,5	195,6
509,9	93,9
513,8	69,7
516,8	56,6
522,4	54,4
535,7	47,1
553,7	49,8
572,7	54,4
$P=98\cdot 10^5$ Pa	
288,7	168,4
311,2	159,9
338,1	151,2
359,0	142,4
391,0	129,1
423,0	115,8
463,9	103,2
488,5	95,9
494,3	96,5
498,3	96,9
502,4	95,6
508,5	95,9
513,8	89,9
522,4	88,8
535,7	84,2
553,7	83,5
572,7	81,4
$P=147\cdot 10^5$ Pa	
288,7	171,1
311,2	163,9
338,1	152,9
359,0	145,5
391,0	132,6
423,0	120,8
463,9	107,8
488,5	101,6
498,3	100,3
502,4	100,3
508,5	99,3
513,8	95,1
522,4	94,5
535,7	92,9
553,7	90,4
572,7	89,4
$P=196\cdot 10^5$ Pa	
288,7	172,0
311,2	166,7
338,1	156,9
359,0	148,1
391,0	136,7
423,0	126,0
463,9	110,3
488,5	106,8
494,3	106,1
498,3	105,2
502,4	104,7
508,5	104,8
513,8	101,7
522,4	99,5
535,7	97,2
553,7	94,1
572,7	92,9
$P=294\cdot 10^5$ Pa	
288,5	179,6
311,2	169,4
338,1	158,5
359,0	153,0
391,0	139,5
423,0	130,1
463,9	119,8
488,5	110,5
494,3	112,6
498,3	109,4
502,4	108,9
508,5	109,4
513,8	106,8
522,4	104,2
535,7	102,1
553,7	100,7
572,7	100,2
$P=392\cdot 10^5$ Pa	
288,7	181,6
311,2	172,1
338,1	164,6
359,0	156,0
391,0	143,9
423,0	133,6
463,9	122,9
488,5	115,9
493,3	115,6
498,3	113,0
513,8	111,5
522,4	109,4
535,7	105,8
553,7	106,8
572,7	105,5
$P=490\cdot 10^5$ Pa	
288,7	185,2
311,2	179,1
338,1	167,3
359,0	159,9
391,0	147,6
423,0	138,3
463,9	125,4
488,5	118,5
494,3	119,5
498,3	117,9
513,8	115,8
522,4	114,5
535,7	112,8
553,7	110,6
572,7	108,9
Methylethylketone	
$P=0{,}98\cdot 10^5$ Pa	
285,9	152,0
321,1	140,4
345,4	129,9
$P=39{,}2\cdot 10^5$ Pa.	
285,9	155,3
321,1	144,2
345,4	132,2
375,9	117,7
401,8	108,6
435,6	101,9
467,5	91,6
470,4	89,6
500,0	88,6
516,2	87,9
523,0	87,0
526,7	89,3
527,8	88,5
530,4	99,6
530,5	123,9
532,9	157,6
535,6	61,5
541,2	62,6
551,3	55,4
564,1	53,6
526,7	112,3
527,8	108,1
530,4	109,7
532,9	110,4
564,1	107,9
578,5	107,0
595,7	107,0
578,5	54,5
595,7	54,5
613,5	57,5
634,6	58,8
$P=98\cdot 10^5$ Pa	
285,9	160,5
321,1	147,2
345,4	135,5
375,9	121,7
401,8	114,7
435,6	104,7
467,5	93,9
470,4	96,5
500,0	94,7
516,2	90,7
523,0	91,5
526,7	90,6
527,8	90,5
530,4	90,5
532,9	90,2
551,3	86,1
578,5	85,0
595,7	84,9
613,5	83,7
634,6	81,9
613,5	109,3
634,6	105,5
$P=490\cdot 10^5$ Pa	
285,9	172,6
321,1	160,9
345,4	149,7
$P=196\cdot 10^5$ Pa	
285,9	162,9
321,1	151,4
345,4	141,4
375,9	123,7
401,8	122,1
435,6	110,4
467,5	102,5
470,4	102,3
500,0	101,3
516,2	98,7
527,8	98,8
530,4	97,0
532,9	97,7
551,3	93,1
578,5	93,0
595,7	93,9
613,5	94,1
634,6	93,9
$P=294\cdot 10^5$ Pa	
285,9	165,5
321,1	153,5
345,4	143,7
375,9	130,2
375,9	138,1
401,8	136,5
435,6	124,2
467,5	119,1
500,0	117,2
516,2	116,2
523,0	117,3
401,8	126,5
435,6	115,9
467,5	107,9
500,0	106,2
516,2	106,0
523,0	106,0
526,7	102,6
527,8	103,2
530,4	104,5
532,9	102,3
564,1	100,9
578,5	100,2
595,7	100,2
613,5	102,3
634,6	98,6
$P=392\cdot 10^5$ Pa	
285,9	169,2
321,1	159,5
345,4	147,4
375,9	133,2
401,8	129,1
435,6	120,4
467,5	113,9
500,0	114,7
516,2	112,1
523,0	112,6
526,7	119,5
527,8	115,1
530,4	114,4
532,9	116,8
564,1	112,6
595,7	113,4
613,5	113,9
634,6	114,2

TABLE 1 (continued)

Methylbutylketone

T, K	λ	T, K	λ	T, K	λ	T, K	λ
$P=0{,}98\cdot 10^5$ Pa		$P=98\cdot 10^5$ Pa		555,2	107,3	417,4	141,6
296,2	140,4	296,2	149,8	601,3	103,2	440,2	135,2
325,9	134,5	325,9	145,4	632,6	101,1	470,4	127,9
358,9	126,0	358,9	139,9	663,3	102,8	505,2	124,1
389,2	117,2	389,2	129,5	693,2	100,1	533,9	123,1
$P=31{,}5\cdot 10^5$ Pa		417,4	123,8	$P=294\cdot 10^5$ Pa		555,2	120,2
296,2	146,3	440,2	117,8	296,2	158,8	601,3	120,1
325,9	138,3	470,4	112,5	325,9	153,4	632,6	118,6
358,9	136,9	505,2	105,9	358,9	147,6	663,9	116,5
389,2	127,1	533,9	101,8	389,2	139,6	693,2	115,5
417,4	121,0	555,2	99,6	417,4	134,6	$P=490\cdot 10^5$ Pa	
440,2	113,5	601,3	95,0	440,2	127,2	296,2	167,2
470,4	109,7	632,6	92,7	470,4	122,6	325,9	161,9
505,2	102,8	663,3	93,5	505,2	120,8	358,9	157,0
533,9	97,3	693,2	93,2	533,9	115,9	389,2	149,9
555,2	94,1	$P=196\cdot 10^5$ Pa		555,2	115,6	417,4	143,6
563,2	106,1	296,2	154,8	601,3	111,0	440,2	139,2
569,4	110,5	325,9	147,8	632,3	109,7	470,4	135,7
574,2	91,1	358,9	142,1	663,3	108,7	505,2	131,4
601,3	72,6	389,2	135,4	693,2	108,3	533,9	128,0
632,6	71,6	417,4	128,3	$P=392\cdot 10^5$ Pa		555,2	126,0
632,6	70,6	440,2	122,5	296,2	163,1	601,3	123,8
663,3	74,4	470,4	116,9	325,9	157,7	632,6	121,6
693,2	75,6	505,2	112,9	358,9	153,1	663,3	120,4
		533,9	110,0	389,2	143,6	693,2	120,6

Dipropylketone

T, K	λ	T, K	λ	T, K	λ	T, K	λ
$P=0{,}98\cdot 10^5$ Pa		685,2	59,6	503,2	110,2	426,3	137,4
294,7	136,6	723,2	62,0	534,5	102,8	447,5	129,8
324,2	132,9	$P=98\cdot 10^5$ Pa		567,2	98,5	467,1	126,9
350,9	124,5	294,7	147,4	591,9	96,9	503,2	122,9
387,7	115,2	324,2	140,2	618,4	91,9	534,5	114,8
$P=28{,}2\cdot 10^5$ Pa		350,9	135,5	654,1	88,9	567,2	109,5
294,7	141,5	387,7	127,8	$P=294\cdot 10^5$ Pa		591,9	108,7
324,2	135,6	426,3	119,8	294,7	158,4	618,4	103,0
350,9	125,4	447,5	114,0	324,2	151,5	654,1	97,4
387,7	122,8	467,1	111,3	350,9	144,8	$P=490\cdot 10^5$ Pa	
426,3	109,9	503,2	104,8	387,7	137,2	294,7	165,3
447,5	103,6	534,5	97,8	426,3	131,5	324,2	159,9
467,1	101,6	567,2	94,8	447,5	125,0	350,9	156,0
503,2	95,5	591,9	93,7	467,1	122,8	387,7	148,8
534,5	89,7	618,4	88,0	503,2	114,6	426,3	142,0
567,2	91,3	654,1	83,9	534,5	108,9	447,5	134,9
591,9	94,9	$P=196\cdot 10^5$ Pa		567,2	103,3	467,1	134,2
599,7	130,2	296,7	151,0	591,9	102,2	503,2	126,2
603,9	132,2	324,2	145,2	618,4	97,5	534,5	120,9
609,2	146,2	350,9	141,5	654,1	92,2	567,2	115,8
610,5	120,2	387,7	132,0	$P=392\cdot 10^5$ Pa		591,9	114,2
613,2	85,1	426,3	125,2	294,7	160,5	618,4	109,0
618,4	59,9	447,5	117,8	324,2	155,5	654,1	102,9
654,1	58,3	467,1	113,8	350,9	148,1		
				387,7	141,3		

where λ_T is the thermal conductivity coefficient at temperature T; λ_1, thermal conductivity coefficient at temperature $0.6T_{cr}$; T_{cr}, the critical temperature. For all the substances studied $0.6T_{cr} < T_b$ (the boiling point). This value was chosen for λ_1 because this point lies in the temperature range in which the thermal conductivity coefficient of liquid ketones has been measured at atmospheric pressure.

The literature offers practically no information on the critical values of ketone parameters. Therefore, it was necessary to calculate the critical temperature and pressure by the Lidersen method [11].

Testing of Eq. (1) for the liquid and gaseous ketones studied revealed that it describes the temperature dependence of thermal conductivity of these substances qualitatively and quantitatively to an accuracy of 1-3%.

TABLE 2. Values of $(\partial\lambda/\partial T)_P \cdot 10^4$, W/m•deg²K and $(\partial\lambda/\partial P)_T \cdot 10^9$, W/m•deg K•Pa for Various Ketones

T, K	Dimethyl		Methylethyl		Methylbutyl		Dipropyl	
	P, 10⁵ Pa							
	196	392	196	392	196	392	196	392
	$\left(\frac{\partial\lambda}{\partial T}\right)_P \cdot 10^4$							
293	−3,75	−3,50	−3,75	−3,25	−2,25	−2,00	−2,50	−2,00
393	−3,50	−3,25	−3,13	−2,88	−2,00	−1,95	−2,13	−1,88
493	−2,25	−2,00	−1,38	−1,05	−1,63	−1,13	−2,00	−1,75
	$\left(\frac{\partial\lambda}{\partial P}\right)_T \cdot 10^9$							
293	0,36	0,26	0,33	0,31	0,48	0,36	0,51	0,46
393	0,61	0,38	0,56	0,46	0,54	0,51	0,56	0,54
493	0,87	0,41	0,64	0,59	0,64	0,61	0,59	0,56

The degree to which Eq. (1) is satisfied for the liquid ketones studied is shown in Fig. 2, whence it is evident that the experimental data fit the straight line well. The equation of this straight line has the form

$$\lambda_T = \left(1.774 - 1.293\frac{T}{T_{cr}}\right)\lambda_1 \quad (2)$$

The relationship between λ_T and boiling point was also established for the liquid ketones:

$$\lambda_T = \left(1,867 - 0,947\frac{T}{T_b}\right)\lambda_{0,9T_b}, \quad (3)$$

where $\lambda_{0.9T_b}$ is the thermal conductivity at $0.9T_b$.

Equations (2), (3) may be used to calculate the thermal conductivity of liquid ketones to an accuracy of 1-3% if the values of λ_1 and $\lambda_{0.9T_b}$ are known.

Equations of the form of Eqs. (2), (3) have also been obtained for acetates, aromatic hydrocarbons, and alkanes [10]. These substances belong to different homological series and their thermal conductivities differ greatly. The above confirms that the law of corresponding states is satisfied well by various homological series of organic compounds.

It would be of interest to relate the quantities λ_1 and $\lambda_{0.9T_b}$ of Eqs. (2), (3) to the molecular mass, boiling point, and critical temperature of the substances. According to Fig. 3, the dependence of λ_1 on molecular mass μ is described by the equation

$$\lambda_1 = \frac{\mu}{9,84\mu - 202}, \ \frac{\text{W}}{\text{m}\cdot\text{K}}. \quad (4)$$

The dependence of $\lambda_{0.9T_b}$ on μ, T_b and λ_1 on T_{cr} are described by the equations

$$\lambda_{0,9T_b} = \frac{\mu}{10,74\mu - 261,6}, \ \frac{\text{W}}{\text{m}\cdot\text{K}}, \quad (5)$$

$$\lambda_{0,9T_b} = \frac{T_b}{22,16T_b - 8066,5}, \ \frac{\text{W}}{\text{m}\cdot\text{K}}, \quad (6)$$

$$\lambda_1 = \frac{T_{cr}}{19,76T_{cr} - 6806,6}, \ \frac{\text{W}}{\text{m}\cdot\text{K}}. \quad (7)$$

Using Eqs. (2), (3) with consideration of Eqs. (4)-(7) one can calculate the thermal conductivity of experimentally unstudied liquid ketones over wide temperature ranges at atmospheric pressure with an accuracy of not less than 3%. These equations have the desired feature that calculations require no preliminary information on liquid ketone thermal conductivity.

By using Eq. (1) the following expression was obtained for the temperature dependence of thermal conductivity in gaseous ketones:

$$\lambda_T = \left(1,72\frac{T}{T_{cr}} - 0,718\right)\lambda_{T_{cr}}. \quad (8)$$

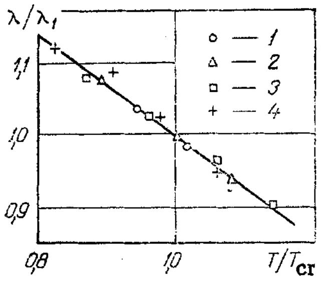

Fig. 2

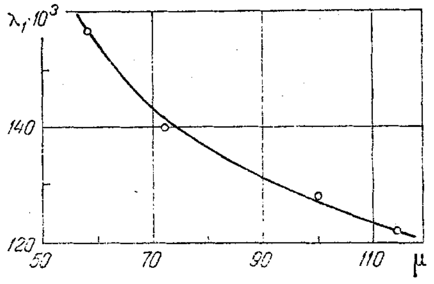

Fig. 3

Fig. 2. Relative thermal conductivity versus relative temperature for liquid ketones: 1) dimethylketone; 2) methylethylketone; 3) methylbutylketone; 4) dipropylketone.

Fig. 3. Quantity λ_1 versus μ for liquid ketones, μ, kg/mole.

where $\lambda_{T_{cr}}$ is the thermal conductivity at T_{cr}. To an accuracy of 1-3% Eq. (8) describes the temperature dependence of thermal conductivity of gaseous ketones over the temperature interval 293-715°K.

Equation (8) has also been obtained for gaseous simple ethers [12]. Simple ethers and ketones belong to differing homological series and their thermal conductivities differ greatly. This again confirms that the law of corresponding states is satisfied well by gaseous organic compounds of different homological series.

For gaseous ketones the dependence of $\lambda_{T_{cr}}$ on T_{cr} was found to be of the following form:

$$\lambda_{T_{cr}} = (18{,}448\cdot10^{-3}T_{cr}^2 + 56{,}248T_{cr} - 3{,}53)\cdot10^{-6},\ \frac{W}{m\cdot K} \tag{9}$$

It follows from Eqs. (8), (9) that

$$\lambda_T = \left(1.72\frac{T}{T_{cr}} - 0.718\right)(18{,}448\cdot10^{-3}T_{cr}^2 + 56{,}248T_{cr} - 3{,}53)\cdot10^{-6},\ \frac{W}{m\cdot K}\ . \tag{10}$$

Equation (10) describes the temperature dependence of thermal conductivity of gaseous ketones to an accuracy of 1-5%. With the aid of Eq. (10) one can calculate the thermal conductivity of experimentally unstudied gaseous ketones as a function of temperature. Such a calculation requires knowledge only of the critical temperature T_{cr}.

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DETERMINATION OF THERMOPHYSICAL CHARACTERISTICS OF MULTICOMPONENT LIQUIDS

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A new approach is developed for determining the specific isobaric heat capacity and thermal expansion coefficient of multicomponent liquids.

Modern techniques of measuring heat capacity are based on the calorimetric method or a method depending on the unique features of sound or ultrasound propagation through the liquid under study [1-5]. For well-known reasons neither of these methods permits determination of the heat capacity of multicomponent "coarse" liquids such as heavy petroleum, suspensions, solutions, etc. with sufficient accuracy. At the present time no methods are known for determination of c_P of such liquids over wide ranges of temperature and pressure, especially at high pressures.

We will consider the possibility of determining the isobaric specific heat of multicomponent liquids commencing from the fact that with adiabatic increase (or decrease) in pressure in a thermostratified liquid-filled vessel there will be a corresponding increase (or decrease) in temperature of the liquid, characterized, in particular, by its specific heat.

The change in liquid temperature under adiabatic compression (or expansion) can be determined theoretically from the thermodynamic relationship [6]

$$\frac{1}{V} TdS = c_P\, dT - T\, \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_P dP. \tag{1}$$

Considering that for a reversible adiabatic process the change in entropy dS is equal to zero, and taking $1/V(\partial V/\partial T)_P$ constant, for a finite pressure change we obtain the following expression for specific (per unit volume) isobaric heat capacity:

$$c_P = \frac{\alpha_P\,(P - P_0)}{\ln(T/T_0)}, \tag{2}$$

where T_0, P_0 and T, P are the initial and final temperature and pressure; $\alpha_P = 1/V(\partial V/\partial T)_P$ is the thermal expansion coefficient at constant pressure. It is insured that the process occurs adiabatically by changing the pressure at a rate such that heat exchange between the system under consideration (thermosensor-liquid shell) and the surrounding thermodynamic medium does not have time to develop.

It has been shown experimentally [7] that the dependence of the temperature change ΔT of a liquid under compression or expansion on the rate of pressure change $\Delta P/\Delta\tau$ is of an exponential nature. Data from experiments with the apparatus to be described below were used to construct $\Delta T = f(\Delta P/\Delta\tau)$ for various liquids at $\Delta P = 10$ MPa (Fig. 1). It is evident that for each liquid the value of $(\Delta P/\Delta\tau)_a$ at which the process can be considered adiabatic differs. We note that of the liquids considered the required rate is highest for kerosene.

The value of $(\Delta P/\Delta\tau)_a$ is related to the heat-exchange properties of both the liquid itself, and the container within which it is located. Moreover, experimental results have shown that $(\Delta P/\Delta\tau)_a$ depends linearly upon the value of the pressure change ΔP. Thus, for example, for kerosene at $\Delta P = 10$ MPa $(\Delta P/\Delta\tau)_a = 1$ MPa/sec, at $\Delta P = 1$ MPa the value of $(\Delta P/\Delta\tau)_a$ is ten times smaller, 0.1 MPa/sec. Thus there exists a certain definite adiabatic pres-

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