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FUNDAMENTAL EQUATION OF STATE OF ARGON, SATISFYING THE SCALING HYPOTHESIS AND WORKING IN THE REGION OF HIGH TEMPERATURES AND PRESSURES

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Summary. This paper discusses the problem of describing thermodynamic properties of a substance at high temperatures and pressures on the basis of the fundamental equation of state (FEoS). This FEoS has the following characteristics: it transforms into the virial equation of state in the region of low densities; it is converted into the Berestov equation in the vicinity of the critical point. FEoS testing has been carried out on known thermodynamic properties of argon and has allowed establishing its workspace: by the pressure up to 1000 MPa; by the temperature from the temperature of the triple point to 1200 K. It has been shown that our FEoS can qualitatively correctly describe the thermal surface of argon up to 17 000 K. A comparison of FEoS has been made with some well-known equations of state. When developing FEoS of argon, we have used elements of the similarity theory, which has allowed reducing the number of individual parameters of this FEoS.

1 INTRODUCTION

We investigate a problem of describing the thermophysical properties of substances in a wide range of temperatures and pressures including the critical region. The problem attracts the attention of many researchers [1–24]. In particular, this problem is actual when studying the behavior of substances:

- in the range of highly developed density fluctuations near the critical point;
- at high temperatures and high pressures.

To describe the properties of pure substances at high pressures and high temperatures, the authors of [25–32] have developed a number of fundamental equations of state (FEoS). When describing the liquid behavior in the vicinity of the critical point, we have used previously a number of approaches and developed some equations of state (EoS):

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- scaling EoSs and crossover EoSs in a parametric form and with (the density, the temperature variables) [1, 2, 10, 12, 16, 24];
- FEoS of the virial type [3, 6, 9, 17];
- FEoS [14] based on requirements of the scaling theory (ST) for the critical region [33];
- FEoS [4,5,7,8,11,15,18–20,22,23] converted into a Widom EoS and valid in the vicinity of the critical point.

We have analyzed approaches [1–12, 14–20, 22–24] and have got the following results. Scaling EoSs [1, 2, 16, 24] and crossover EoSs [10, 12] meet the requirements of ST [33], but they have a narrow work area limited by temperatures T ($0.9T_c < T < 2T_c$ [12], here T_c is the critical temperature), and, therefore, can not be used when modeling thermodynamic properties of a substance in the range of high temperatures and high pressures.

One of the disadvantages of the crossover EoS [12] is the need to use different critical temperatures: one T_c to calculate the pressure (p) and another T_c to calculate the isochoric heat capacity (C_V). FEoS [3, 6, 9, 17] do not meet the requirements of ST. Therefore, these EFoSs do not describe the sound velocity (w), C_V, the isobar heat capacity (C_p), and isothermal compressibility factor (K) in the critical region with acceptably small uncertainties. At the same time, these EFoSs describe the equilibrium properties of argon in the regular part of the thermodynamic surface with low uncertainties. For example, FEoS of argon is proposed [9] in this form. The workspace of FEoS FEoS [9] is (limited by pressures $0 \leqslant p \leqslant 1000$ MPa, by temperatures $83 \leqslant T \leqslant 700$ K) and can be successfully used when predicting thermal properties at high temperatures.

Bezverhiy *et al* [14] has developed FEoS, which takes into account the feature presence of $C_V(T,\rho)$ as a known function in the critical region, here ρ is the density. Our analysis shows that EFoS [14] reproduces power laws of ST qualitatively incorrectly. For example, the critical isotherm [14] follows $\Delta p \propto (\Delta\rho)^3$. It should be $\Delta p \propto \Delta\rho|\Delta\rho|^{\delta-1}$ [33], here $\Delta p = (p-p_c)/p_c$; $\Delta\rho = (\rho-\rho_c)/\rho_c$; p_c is the critical pressure; ρ_c is the critical density; δ is the critical index of the critical isotherm.

Empirical FeoSs are proposed in [5,7,8,11]. They qualitatively correctly reproduce all of ST power laws when describing properties in the vicinity of the critical point.

On the basis of the phenomenological theory of the critical point [34], the authors of [4, 15, 18–20, 22, 23] developed FEoS which is not inferior to scaling EoS and crossover EoS when describing the asymptotic vicinity of the critical point. We mark that FEoSs [4, 5, 7, 8, 11, 15, 18–20, 22, 23] do not satisfy the theory of extended scaling [2]. Indeed, the function, $C_V(T,\rho_c)$ [4, 5, 7, 8, 11, 14, 15, 18–20, 22, 23], follows $C_V \simeq A\tau^{-\alpha} + C\tau$. It is shown in [2] that

$C_V(T,\rho_c)$ should be $C_V \simeq A\tau^{-\alpha} + B\tau^{-\alpha+\Delta} + C\tau$, here $\tau = (T - T_c)/T_c$; α and Δ are the critical indexes. In addition, our analysis shows that FEoSs [4, 5, 7, 8, 11, 14, 15, 18–20, 22, 23] are inferior to FEoS [9] when describing properties at high temperature region. For example, there are a discrepancy between p values calculated by EFoS [9] and p values calculated by [20] at 17 000 K these deviations exceed 50%.

In this paper on the basis of the approach [35], we plan to develop a FEoS that meets the following requirements:

- satisfies ST requirements [33] and does not inferior to EoSs [10, 12] when describing properties in the critical region;
- simulates the thermal surface of argon at temperatures up to 17 000 K and by pressures up to 12 GPa;
- can be converted into the Berestov equation [2] in the critical region.

2 STRUCTURE OF FEOS

By analogy with [4, 15, 18–20, 22, 23], this FEoS has the following structure:

$$F(\rho,T) = F_{\text{reg}}(\rho,T) + F_{\text{nreg}}(\rho,T), \tag{1}$$

where $F(\rho,T)$ is the Helmholtz free energy; $F_{\text{reg}}(\rho,T)$ is a regular function; $F_{\text{nreg}}(\rho,T)$ is an irregular component of the Helmholtz free energy:

$$F_{\text{nreg}}(\rho,T) = RT_c\phi(\omega,t)\left(|\Delta\rho|^{\delta+1}a_0(x) + |\Delta\rho|^{\delta+1+\frac{\Delta}{\beta}}a_1(x)\right), \tag{2}$$

where $\phi(\omega,t)$ is the regular function; R is the gas constant; $\omega = \rho/\rho_c$; $x = \tau/|\Delta\rho|^{1/\beta}$ is the scaling variable; $t = T/T_c$; β is the critical index.

We notice: there is a principal difference of our FEoS from FEoS [4, 15, 18–20, 22, 23]. We have included an additional component in $F_{\text{nreg}}(\rho,T)$. There is a special scale function, $a_1(x)$, in this additional component recommended in [35]. This principal modernization has allowed us to improve a FEoS structure and to meet the requirements [2].

The scaling functions $a_0(x)$ and $a_1(x)$ are calculated based on the following:

- a new representation of the scaling hypothesis [34, 35];
- the Benedek hypothesis [36];
- the Berestov equation [2].

i	$j=0$	$j=1$	$j=2$
0	0	0	2.727 031 612 1447
1	0	0	−2.180 917 085 2935
2	0	0	2.018 179 285 6405
3	0	−1.651 807 350 2083	1.951 547 138 476
4	0	3.235 0 97 0 27 9 45 2	5.861 967 866 4433
5	0	0.203 261 164 281 07	−2.078 078 708 984
6	−0.283 648 592 739 017	−1.850 670 154 3516	−2.801 735 606 0172
	−0.031 673 399 139 638	2.497 843 489 6566	9.620 211 455 1673
	−0.117 319 511 789 66	−1.164 995 874 2581	−3.476 213 158 3227
9	0.413 193 730 791 89	2.810 166 015 2324	−0.811 288 614 251 57
10	− 0.765 606 737 657 49	−2.801 124 973 5011	0.766 664 260 646 57
11	− 0.650 491 354 2378	−2.592 742 798 4863	0.450 684 903 397 98
12	1.808 588 644 5017	5.578 734 268 4796	−0.239 259 405 790 52
13	−1.042 305 956 028	−2.485 069 647 1961	−1.024 947 033 0846
14	−0.813 944 971 192 75	−2.392 997 971 8019	0.759 164 862 584 33
15	1.328 057 607 1621	3.119 794 125 8801	0
16	−0.486 803 106 500 06	−0.933 985 969 4002	−0.221 489 078 823 57
17	−0.243 474 625 433 64	−0.585 625 628 796 48	0.091 451 137 589 177
18	0.327 530 667 992 16	0.660 860 378 937 56	0
19	−0.154 068 046 320 52	−0.286 546 955 475 54	−0.008 667 473 663 7731
20	0.039 124 504 337 479	0.068 175 553 922 501	0.001 865 695 143 8862
21	−0.005 346 887 409 843	−0.008 772 962 958 1014	0
22	0.000 310 067 180 058 02	0.000 478 732 897 948 04	$-2.5448089017224 \times 10^{-5}$

Table 1: Coefficients $C_{i,j}$ of FEoS (1).

These functions are written the following form:

$$a_0(x) = -\frac{u_0 k \gamma_1 x_0^{2-\alpha}}{2\alpha b^2 \alpha_1 (1-\varepsilon)} \left[(\varphi+\varphi_1)^{2-\alpha} - \varepsilon(\varphi+\varphi_2)^{2-\alpha} \right] + \frac{u_0 x_0^{\gamma}}{2k} (\varphi+\varphi_3)^{\gamma} + u_0 C_0, \quad (3)$$

$$a_1(x) = -\frac{u_1 k \gamma_2 x_0^{2-\alpha+\Delta}}{2\alpha b^2 \alpha_2 (1-\varepsilon)} \left[(\varphi+\varphi_1)^{2-\alpha+\Delta} - \varepsilon(\varphi+\varphi_2)^{2-\alpha+\Delta} \right] + \frac{u_1 x_0^{\gamma+\Delta}}{2k} (\varphi+\varphi_3)^{\gamma+\Delta} + u_1 C_1, \quad (4)$$

where $\varepsilon_0 = x_1/x_2$; $\alpha_1 = (2-\alpha)(1-\alpha)$; $\gamma_1 = \gamma(\gamma-1)$; $\gamma_2 = (\gamma+\Delta)(\gamma+\Delta-1)$; $\alpha_2 = (2-\alpha+\Delta)(1-\alpha+\Delta)$; $\varphi = x/x_0$; $\varphi_i = x_i/x_0$, $i \in \{1,2,3\}$; $b^2 = (\gamma-2\beta)/[\gamma(1-2\beta)]$; $k = [(b^2-1)/x_0]^{\beta}$; γ is the critical index; x_0, u_0 and u_1 are the individual parameters; $Z_c = p_c/(R\rho_c T_c) \times 10^3$; C_0

i	$j=3$	$j=4$	$j=5$
0	4.482 248 574 7539	2.332 643 055 2399	1.808 465 772 8776
1	−3.225 639 106 0006	−1.048 810 609 669	−0.932 531 831 731 91
2	−3.143 085 800 7921	−7.416 650 230 6154	−4.156 240 517 2991
3	7.651 653 302 7528	6.805 576 926 7176	3.940 459 100 9914
4	6.118 667 623 2535	4.737 381 837 8476	0.741 441 138 784 28
5	−8.029 797 760 4914 6	−8.232 510 077 0624	−2.172 672 507 2028
6	−0.081 651 952 400 293	1.488 772 709 3593	0.930 235 432 967 88
7	8.388 156 700 3335	2.510 816 288 7711	−0.130 873 463 355 37
8	−4.059 017 137 1799	−1.089 247 218 7001	0
9	−0.103 759 991 449 93	−0.277 754 425 60302	0
10	1.129 529 996 898	0.093 168 194 589 203	0
11	−0.543 765 088 527 54	0.151 336 027 7963	0
12	0.008 645 103 248 2461	−0.082 484 913 633 882	0
13	0.001 821 779 428 3432	0.012 151 299 548 948	0

Table 2: Coefficients $C_{i,j}$ of FEoS (1).

and C_1 are the constant coefficients which value is found from the equations

$$(\delta+1)a_0|_{\varphi=-1}+\frac{x_0}{\beta}a'_0|_{\varphi=-1}=0, \tag{5}$$

$$\left(\delta+1+\frac{\Delta}{\beta}\right)a_1|_{\varphi=-1}+\frac{x_0}{\beta}a'_1|_{\varphi=-1}=0. \tag{6}$$

We have selected the regular component (1) in the form [19]:

$$\begin{aligned}F_{\rm reg}(\rho,T)&=F^0(\rho,T)+RT\omega y_2+RT\omega(Z_c-0.2)y_6+RT\omega D_3(y_4-y_6)\\&+RT\omega\tau_1\Big[D_1(\omega-3)+D_2\big(\omega^2-2\omega\big)\Big]+RT\omega\sum_{i=0}^{22}\sum_{j=0}^{20}\big(C_{i,j}\tau_1^j\Delta\rho^i\big),\end{aligned} \tag{7}$$

where $F^0(\rho,T)$ is the ideal gas component of $F(\rho,T)$; $\tau_1=T_c/T-1$; functions y_2, y_4, y_6 have the following form: $y_2=-7.7/6+2.9/6\Delta\rho-1.1/6\Delta\rho^2+0.05\Delta\rho^3$, $y_4=5-4\Delta\rho+3\Delta\rho^2-2\Delta\rho^3+\Delta\rho^4$, $y_6=4-3\Delta\rho+2\Delta\rho^2-\Delta\rho^3+\Delta\rho^5$.

We have calculated φ_1, φ_2, φ_3 values according to the method detailed in [15]. It let us got $\varphi_1=2.807\,223\,47$, $\varphi_2=14.471\,7304$, $\varphi_3=5.732\,468\,25$.

i	$j=6$	$j=7$	$j=8$
0	1.942 056 320 0621	3.245 246 493 1065	−8.239 406 700 9885
1	−1.034 640 564 3285	−0.970 159 560 317 12	−0.011 163 693 637 208
2	−0.944 139 567 2871	0.298 859 602 686 75	0.039 252 086 979 538
3	1.204 115 946 6534	0	0
4	−0.327 499 512 264 19	0	0
i	$j=9$	$j=10$	$j=11$
0	−18.746 448 404 883	51.077 633 966 366	68.645 329 452 91
1	0.541 072 550 799 12	0	0
2	−0.178 044 619 880 26	0	0
i	$j=12$	$j=13$	$j=14$
0	−182.047 371 3271	−144.870 071 874 34	383.406 155 478 06
i	$j=15$	$j=16$	$j=17$
0	174.017 641 515 55	−472.418 838 330 36	−110.347 173 018 13
i	$j=18$	$j=19$	$j=20$
0	314.552 869 844 35	28.506 239 206 301	−87.384 487 306 415

Table 3: Coefficients $C_{i,j}$ of FEoS (1).

We have chosen the crossover function in accordance with the recommendations [7]:

$$\phi(\omega,t)=\phi_0(\omega)\phi_1(t),\qquad \phi_0(\omega)=\Big[\big(1-\omega\big)^m-1\Big]^2,\qquad \phi_1(t)=1/t^2, \tag{8}$$

where $m\in N$.

We have tested FEoS (1) with components (2)–(4) and (7) on the example describing the equilibrium properties of argon [37–54].

3 FEOS OF ARGON

Select the ideally-gas component of argon $F^0(T,\rho)$ according to the recommendations of [9]:

$$F^0(\rho,T)=RT\big(\ln\omega+a_1^0+a_2^0t^{-1}-1.5\ln t\big), \tag{9}$$

where $a_1^0=58.316\,662\,43$ and $a_2^0=524.946\,511\,64$.

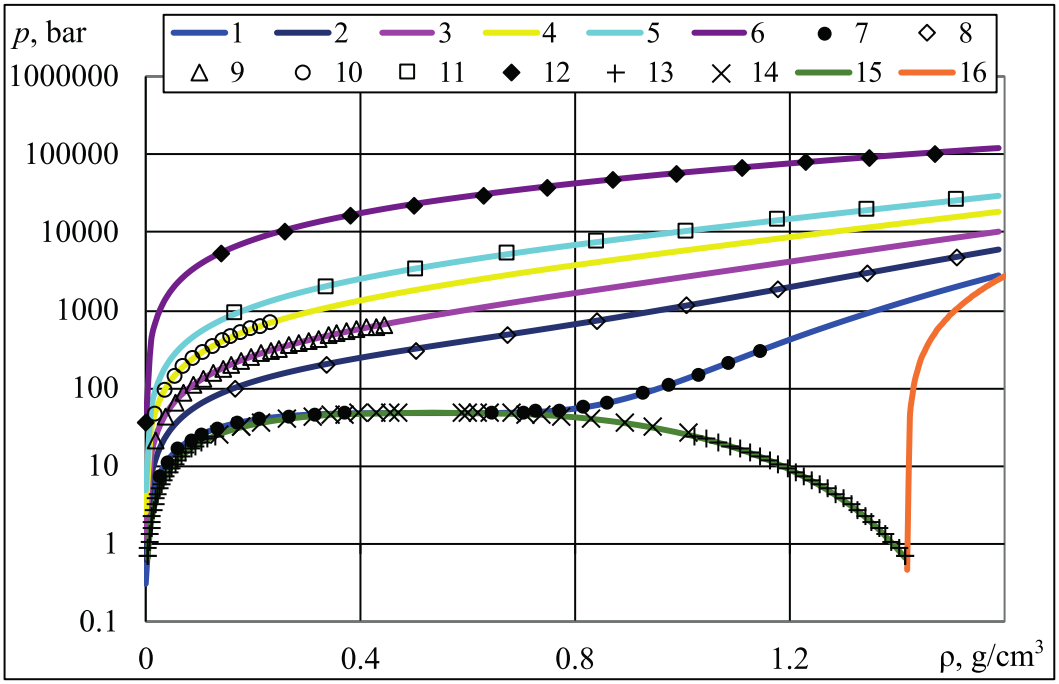

Figure 1: Isotherms, the liquid–gas coexistence curve and the melting line of argon. Isotherms calculated using FEoS (1): 1—critical isotherm; 2—300 K; 3—573.15 K; 4—1223.15 K; 5—2300 K; 6—17 000 K. Experimental data: 7—150.65 K [42]; 8—300 K [3]; 11—2300 K [3]. Tabulated data: 9—573.15 K [43]; 10—1223.15 K [43]; 12—17 000 K [9]; 13—tabulated data on the density at the saturation line [9]; 14—experimental data on the density of a saturated liquid and saturated vapor [49]; 15—data on the density at the saturation line calculated by FEoS (1); 16—data on the density at the melting line [9].

We have calculated expressions for compressibility Z on the basis of FEoS (1) with components (2)–(4) and (7):

$$\begin{aligned} Z(\rho,T) = {} & 1 + y_1\omega^2 + y_2\omega + D_3\left(y_3\omega^2 + y_4\omega - y_5\omega^2 - y_6\omega\right) + \left(y_5\omega^2 + y_6\omega\right)(Z_c - 0.2) \\ & + \omega\sum_{i=0}^{22}\sum_{j=0}^{20} C_{i,j}{\tau_1}^j\Delta\rho^{i-1}(i\omega + \Delta\rho) + D_1\omega\tau_1(2\omega - 3) + D_2\omega^2\tau_1(3\omega - 4) \\ & + Z_c\omega|\Delta\rho|^{\delta}\phi_1(t)t\left(\phi_0(\omega)\mathrm{sign}(\Delta\rho)h_0(x) + {\phi'}_0(\omega)|\Delta\rho|a_0(x)\right) \\ & + Z_c\omega|\Delta\rho|^{\delta+\frac{\Delta}{\beta}}\phi_1(t)t\left(\phi_0(\omega)\mathrm{sign}(\Delta\rho)h_1(x) + {\phi'}_0(\omega)|\Delta\rho|a_1(x)\right), \end{aligned} \tag{10}$$

where $y_{2i-1} = {y'}_{2i}(\omega)$ $(i \in \{1,2,3\})$; $h_n(x)$ are scale functions of chemical potential [27]:

$$h_0(x) = (\delta+1)a_0(x) - \frac{x}{\beta}{a'}_0(x), \qquad h_1(x) = \left(\delta + 1 + \frac{\Delta}{\beta}\right)a_1(x) - \frac{x}{\beta}{a'}_1(x). \tag{11}$$

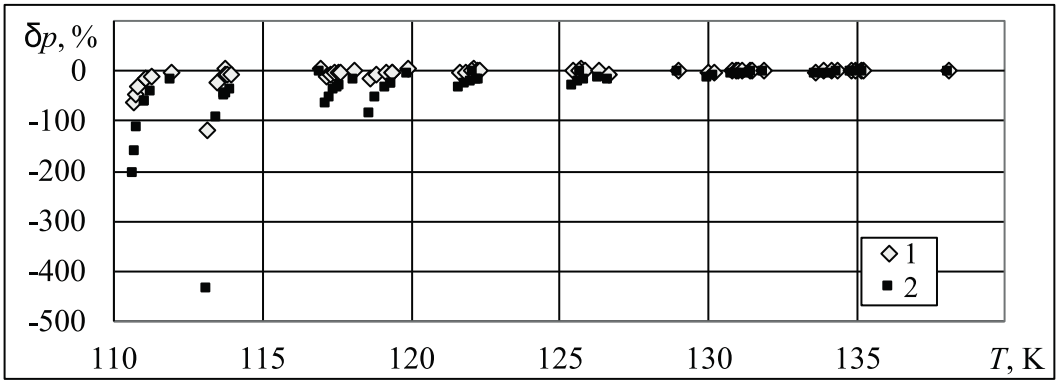

Figure 2: Relative deviations $\delta p = (p_{\text{exp}} - p_{\text{cal}})/p_{\text{exp}}100\%$ corresponded to p_{cal} values calculated with the help of FEoS (1) and FEoS [9] in the metastable states of argon: (p_{exp}, ρ_{exp}, T_{exp}) data are taken from [45] over isochoric lines 1231.9, 1210.9, 1180.2, 1165.6, 1140.9, 1099.8, 1050.8 and 1010.7 kg/m^3; 1—p_{cal} values calculated with the help of FEoS (1); 2—p_{cal} values calculated with the help of FEoS [9]. On each of the isochors, two experimental points corresponding to large values of the pressure are located in the single-phase range, the rest of experimental points are located in the metastable range.

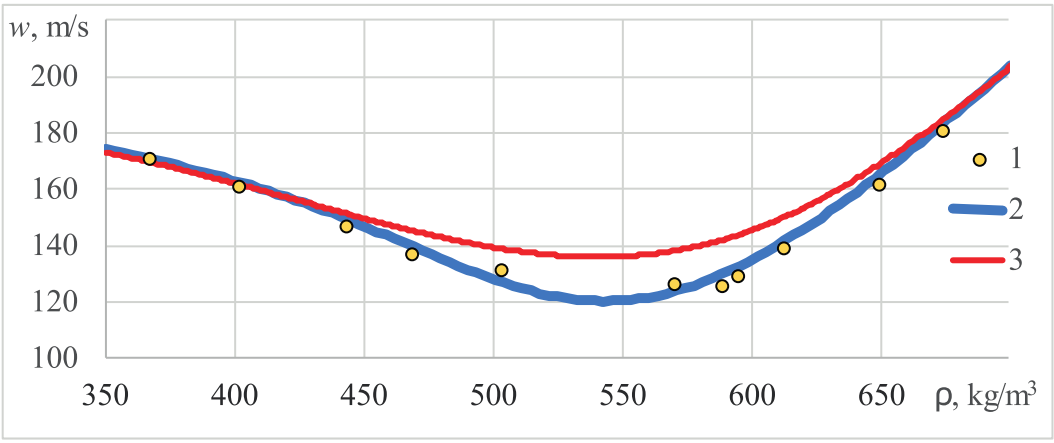

Figure 3: Argon sound speed at 150.8 K isotherm: 1—experimental data [46]; 2—calculation by FEoS (1); 3—calculation by FEoS [9].

Coefficients and parameters of FEoS (1) with components (2)–(4) and (7), (8) were determined on the basis of an array of experimental data [37–54] among them: $T_c = 150.66$ K, $p_c = 4.8634$ MPa, $\rho_c = 535.1$ kg/m^3, $R = 0.208\,133\,32$ J/(g K), $u_0 Z_c = 4.549\,364\,19$, $u_1 Z_c = 0.052\,429\,623\,1552$, $\alpha = 0.11$, $\beta = 0.3255$, $\gamma = 1.239$, $\delta = 4.806$, $\Delta = 0.51$, $m = 3$, $D_1 = 0.528\,541\,695\,546\,02$, $D_2 = 0.874\,668\,218\,972\,52$, $D_3 = -7.913\,173\,555\,7194 \times 10^{-3}$ and $x_0 = 0.311\,220\,376\,399\,66$. The values of coefficients $C_{i,j}$ are presented in tables 1, 2 and 3.

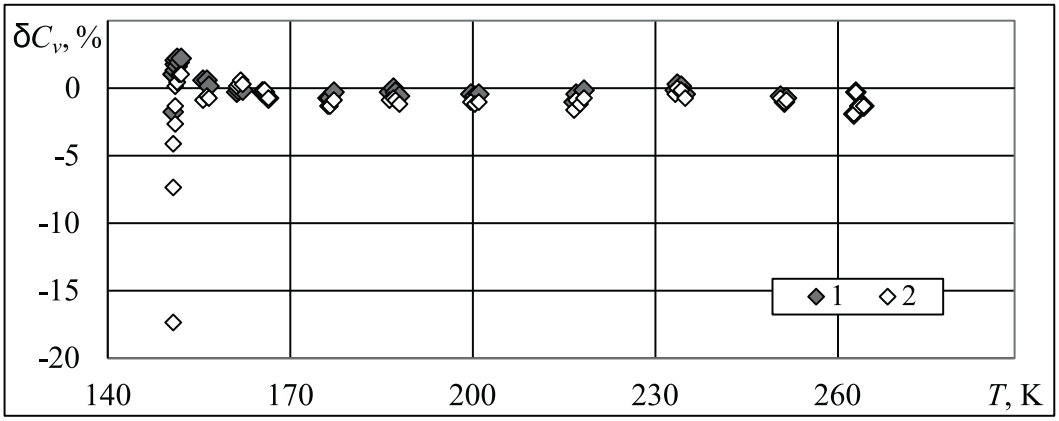

Figure 4: Relative deviations $\delta C_V = (C_{V,\text{exp}} - C_{V,cal})/C_{V,\text{exp}}100\%$; p_{cal}: $C_{V,cal}$ corresponded to values calculated with the help of (1) in the single phase range; $C_{V,\text{exp}}$ corresponded to data [48] over isochoric line 473.6 kg/m^3; 1—C_V calculated with the help of FEoS (1); C_V calculated with the help of FEoS [9].

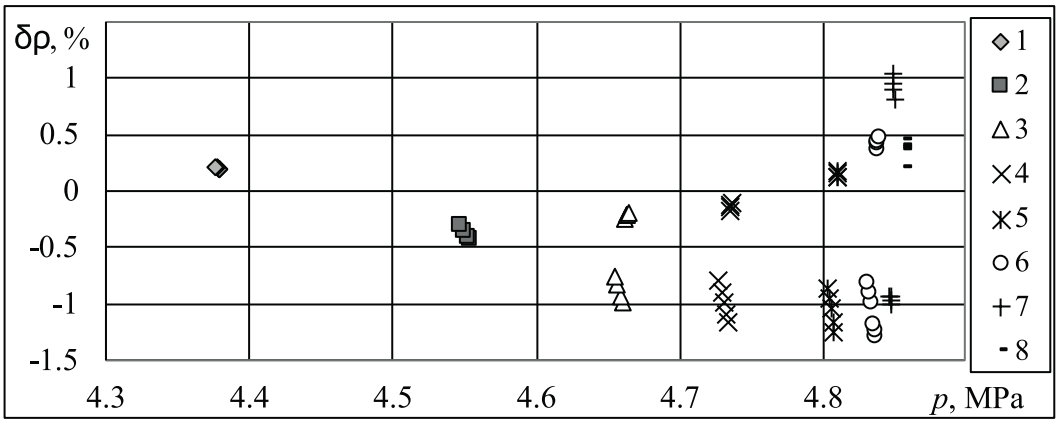

Figure 5: Relative divergence values of density $\delta\rho = (\rho_{\text{exp}} - \rho_{\text{calc}})/\rho_{\text{exp}}100\%$ calculated as per the equations pre-sented in this study as compared with the experimental data [44]. Isothermal lines: 1—148.007 K; 2—149.006 K; 3—149.598 K; 4—149.983 K; 5—150.372 K; 6—150.52 K; 7—150.579 K; 8—150.621 K.

Based on compressibility Z (10), we have calculated the thermal surface of argon (figure 1). As one can see, FEoS (1) transmits the thermal surface of argon in the temperature range from the saturation line and the melting line to 2300 K and it can be extrapolated by temperature up to 17 000 K and by pressure up to 12 GPa. FEoS (1) describes the experimental (p,ρ,T)-data in the metastable range [45], experimental data about C_V [48] and about the speed of sound w [46] in the vicinity of the critical point with less uncertainty than FEoS NIST [9] (figures 2, 3 and 4). Note that when searching for the coefficients of FEoS (1), experimental data [45, 46] were not used. The FEoS (1) represents experimental (p,ρ,T)-data [44] within the range of the experimental error (figure 5). Experimental data on C_V [48, 50] are transmitted within the experimental error (figure 6) in a wide range of state parameters including the vicinity of the critical point.

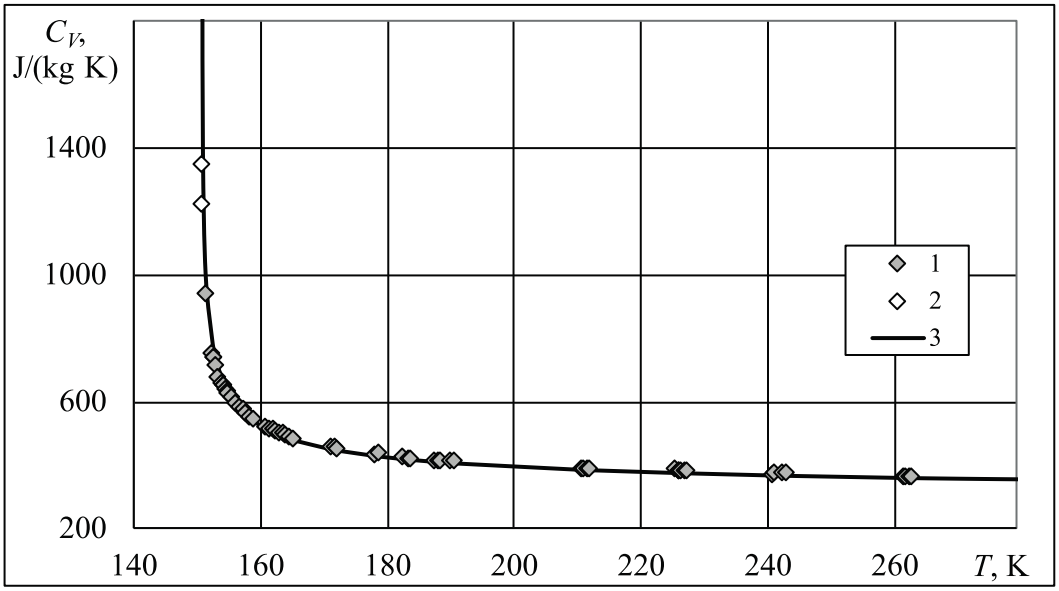

Figure 6: Behavior of the isochoric heat capacity of argon in a single-phase region: 1—experimental points [48] over the isochor of 531 kg/m^3; 2—experimental points [50] over the isochor 530 kg/m^3; 3—C_V calculated with a help of the FEoS (1) over isochor 530.5 kg/m^3.

4 CONCLUSIONS

On the basis of a new representation of the scaling hypothesis [34, 35] and the Berestov equation [2], FEoS (1) was developed. This FEoS primarily works satisfactorily in a wide range of pressures and temperatures including the critical range and the range of high temperatures and pressures. FEoS (1) with components (2)–(4) and (7) has the properties of the virial series in the regular part of the thermodynamic surface as well as the properties of the Berestov equation in the critical range.

Argon FEoS (1) can be used to calculate the equilibrium properties in various technology processes. We have analyzed properties calculated with the help of FEoS (1) in the vicinity of the critical point. Our values significantly exceeds the accuracy of the data generated with the help of known FeoSs and known crossover EoSs [6, 9, 12, 17].

The proposed method of constructing FEoS can be recommended for developing EoSs of substances, which have reliable experimental data, for example, it carbon dioxide and sulfur hexafluoride.

Calculated values of properties from the FEoS (1) to verify computer code are $T = 400$ K, $\rho = 1000$ kg/m^3, $p(T,\rho) = 168\,974.25$ kPa, $C_V(T,\rho) = 0.392\,0699$ J/(g K).

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