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Tao-Mason equation of state for refractory metals

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Abstract

A statistically mechanical-based equation of state has been employed to calculate the liquid density of refractory metals over a wide range of temperatures and pressures. There are three temperature-dependent quantities that are required to use the EOS: the second virial coefficients B_2 , an effective van der Waals co-volume, b and a correction factor, α . The second virial coefficients are calculated from a two-parameter corresponding states correlation, which is constructed with two constants as scaling parameters, i.e., melting temperature T_m and molar density in melting point ρ_m . Our calculations on the liquid density of tantalum, rhenium, molybdenum, titanium, zirconium, hafnium, and niobium from undercooled temperatures up to several hundred degrees above the boiling point (1650 K-7400 K) and pressures ranging from 0 up to 200 MPa reproduces very accurately the experimental PVT data. In order to evaluate the proposed correlation equation for the second virial coefficient, we have compared our results for these systems with those obtained earlier. Our results are in favor of the preference of the TM EOS over two other equation of state.

Keywords: Refractory Metals, Density, Equation of state, Tao- Mason EOS.

Introduction

Refractory metals are extraordinarily resistant to heat and wear. The expression is mostly used in the context of materials science, metallurgy and engineering. The definition of which elements belong to this group differs. The most common definition includes five elements: two of the fifth period (niobium and molybdenum) and three of the sixth period (tantalum, tungsten, and rhenium). They all share some properties, including a melting point above 2000 °C and high hardness at room temperature. They are chemically inert and have a relatively high density. Their high melting points make powder metallurgy the method of choice for fabricating components from these metals. Some of their applications include tools to work metals at high temperatures, wire filaments, casting molds, and chemical reaction vessels in corrosive environments. Partly due to the high melting point, refractory metals are stable against creep deformation to very high temperatures.

The experimental measurements are very difficult due to handling with high temperatures. Static measurements are difficult to perform on most metals because of the typically high values of critical temperature and pressure, problems with samplecontainer contamination, and physical strength limits of high- pressure vessels. Therefore, these measurements are typically carried out using dynamic techniques due to the problems inherent in performing the measurements statically. Because of difficulties in experimental measurements, accurate predictions using theoretical methods are of special importance.

The description of properties of fluids using accurate equation of state is considered as an efficient tool for the calculation of thermodynamic properties (Chapmann *et al.*, 1988; Boublik *et al.*, 1999). In fact, equations of state attempt to describe the relationship between temperature, pressure, and volume for a given substance or mixture of substances. An accurate analytical equation of state for

fluids is a very useful quantity, especially if it has a firm basis in statistical-mechanical theory.

Previously we have successfully extended a statistically mechanical-based equation of state to calculate PVT properties of refrigerant fluid mixtures, LNG, CNG and Ionic liquids (Yousefi *et al.*, 2009; Karimi *et al.*, 2011a; Karimi *et al.*, 2011b; Yousefi & Karimi, 2011). It is the purpose of this work to employ the previous equation of state to predict the liquid density of refractory metals.

Tao-Mason equation of state

Song and Mason proposed an analytical equation of state (SM EOS) in 1989 which was based on statisticalmechanical perturbation theory for hard spheres (Song & Mason, 1989). To derive a density-explicit EOS, they considered Weeks-Chandler-Andersen decomposition of the potential energy (Weeks *et al.*, 1971) as well as the Carnahan-Starling formula for the pair distribution function at contact (Carnahan & Starling, 1969). They developed the SM analytical equation of state to real molecular fluids (Song & Mason, 1990a; Song & Mason, 1990b) by introducing the average contact pair distribution function and tested the resulting EOS against PVT experimental data (Song & Mason, 1990c). The general form of the SM EOS reads as:

$$\frac{P}{\rho KT} = 1 + B_2 \rho + \alpha \rho [G(\eta) - 1] \tag{1}$$

Where P is the pressure, ρ is the number density, kT has the usual meaning; B2 is the second virial coefficient, α is a scaling factor, G(η) is the effective contact pair distribution function, and η is the packing fraction.

Ihm *et al.* (1992) presented and applied a strong principle of corresponding states for non-polar fluids (Ihm *et al.*, 1991; Ihm *et al.*, 1992; Song & Mason, 1992) to the SM EOS. This modification reduced the order of SM EOS from five to three. Although the resulting cubic EOS (hereafter we refer it as ISM EOS) was not accurate in the non-analytical critical and two phase regions, they



claimed that this EOS described the volumetric behavior of non-polar fluids very accurately over the entire range from the dilute gas to the dense liquid. The final form of the ISM EOS reads as:

$$\frac{P}{\rho KT} = 1 - \frac{(\alpha - B_2)\rho}{1 + 0.22\lambda b\rho} + \frac{\alpha\rho}{1 - \lambda b\rho}$$
(2)

Where λ is an adjustable parameter. In this equation b is the effective van der Waals covolume. The ISM equation of state cannot be applied to critical region because its original theory includes a mean filed approximation. Also it cannot be applied to two-phase region to predict vapor pressure of fluids because of the analytic nature of the EOS leads to producing van der Waals loops in the sub-critical P-V isotherms instead of the correct first order vapor liquid transition.

In 1994, Tao and Mason calculated a perturbation correction term for the effect of attractive forces and combined with aforementioned equations of state to present an improved equation of state (TM EOS) (Tao & Mason, 1994). The final form of the TM EOS is:

$$\frac{\rho}{\rho KT} = 1 + (B_2 - \alpha)\rho + \frac{\alpha\rho}{1 - \lambda b\rho} + A_1(\alpha - B_2)b\rho^2 \frac{(e^{\frac{kT_c}{T}} - A_2)}{1 + 1.3(b\rho)^4}$$
(3)

Where

$$A_{1} = 0.143$$

$$A_{2} = 1.64 + 2.65[e^{(k-1.093)} - 1]$$
(4)

 $k = 1.093 + 0.26[(\omega + 0.002)^{\frac{1}{2}} + 4.50(\omega + 0.002)]$ (5) Where ω is the Pitzer acentric factor.

Nevertheless, the overall structure of the final equation is quite similar to the previous ones, and gives accurate vapor pressures and improved orthobaric densities of the liquid and vapor from the triple point to near the critical point.

In present work we will utilize the TM EOS (Tao & Mason. 1994) to predict the liquid density of refractory metals, including Ta, Re, Mo, Ti, Zr, Hf, and Nb from undercooled temperatures up to several hundred degrees above the boiling point over a wide pressure range.

Parameters estimation

The Tao-Mason equation of state requires usage of the second virial coefficient B_2 , α and b. If the intermolecular potential is not available, knowledge of experimental second virial coefficient data is sufficient to calculate value of the other two temperature- dependent parameter (Ihm et al., 1991). For refractory metals,

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neither the intermolecular potential nor the experimental B₂ values are available. In this case, there are several schemes. correlation usually based on the corresponding-state principle, by which second virial coefficient can be calculated.

Briefly, the corresponding-states correlations have been based on several factors: a) the critical constants (Tao & Mason, 1992; Tao & Mason, 1994) b) the heat of vaporization and the liquid density at the freezing point (Boushehri & Mason, 1993), c) the surface tension and the liquid density at the freezing point (Ghatee & Boushehri, 1996), d) the boiling points (Mehdipour & Boushehri, 1998), the normal boiling point constants (Eslami, 2001), and e) the speed of sound (Sheikh et al., 2002; Papari et al., 2003). For instance, in their original work, Tao and Mason (1992, 1994) used the macroscopic corresponding-states correlations for B₂ developed by Tsonopoulos (1974) using two arbitrary constants T_c and P_c and the acentric factor ω , to predict density and vapor pressure of simple fluids. They correlated the temperature-dependent parameters $\alpha(T)$ and b(T) in terms of Boyle temperature and Boyle volume (Tao & Mason, 1994). As a result of this, we decided to investigate about a new corresponding states correlation in order that TM EOS could be applied to refractory metals. In this respect, the following correlation equation for B₂ using new scaling parameters, such as the temperature T_m and the molar density ρ_m , both at the melting point has been developed. The resulting correlation for the second virial coefficient reads as follows:

$$B_{2}\rho_{m} = 1.033 - 3.0069 \left(\frac{T_{m}}{T}\right) - 10.588 \left(\frac{T_{m}}{T}\right)^{2} + 13.096 \left(\frac{T_{m}}{T}\right)^{3} - 9.8968 \left(\frac{T_{m}}{T}\right)^{4} \quad (6)$$

$$\alpha\rho_{m} = a_{1}e^{-c_{1}\left(\frac{T}{T_{m}}\right)} + a_{2}[1 - e^{-c_{2}/\left(\frac{T}{T_{m}}\right)}] \quad (7)$$

$$bl_{\ell} = a_{1}\left[1 - c_{1}\left(\frac{T}{T_{m}}\right)\right]e^{-c_{1}\left(\frac{T}{T_{m}}\right)} + a_{2}\left\{1 - \left[1 + \frac{c_{2}}{4\left(\frac{T}{T_{m}}\right)^{\frac{1}{4}}}\right]e^{-\frac{c_{2}}{\left(\frac{T}{T_{m}}\right)^{\frac{1}{4}}}\right\}$$

(8)

Where the constant a_1 , a_2 , c_1 , c_2 are -0.0860, 2.3988, 0.5624, 1.4267, respectively. Knowing the temperaturedependent parameters, the equation of state can be

Table 1. Coefficienst in Eq (14)

1. Metal	2. a	3. b	4. c	5. d	6. e	7. f
8. Ta	9. 73.06	101245.10	11. 8461.60	1227852.55	13. 43978.82	1426701.19
15. Re	1670.96	17. 1576.41	1813563.10	19. 57825.52	20122371.09	21. 102573.72
22. Mo	23. 5.17	247.41	25. 6.24	262.51	27. 0.49	280.04
29. Ti	30. 22836.90	31352352.21	32. 2174137.70	336704879.10	34. 10333677.00	356367083.50
36. Nb	3742.98	38. 1069.46	3910183.32	40. 48227.74	41113640.84	42. 106098.04
43. Zr	44. 1.85	45. 7.30	46153.34	47. 1086.00	483502.78	49. 3727.74
50. Hf	51. 282.58	526000.23	53. 51363.35	54220330.84	55. 473962.54	56409882.57

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Table 3. The calculated results for the saturated liquid density of Ta, Re and Mo at high pressure compared with the experimental data (Jager et al., 1992; Hixon & Winkler, 1992; Thevenin et al., 1993) and other models.

applied to predict the liquid density of fluids over a wide range of temperatures and pressures.

Table 2. Parameters used for refractory metals						
Metal	T _m [K]€	ρ _{m.p} [g/cm³] [€]	T _c [K]€			
Та	3290.15	15.00	16500			
Re	3458.15	18.90	18900			
Мо	2896.15	9.33	1450			
Ti	1941.15	4.11	5850			
Nb	2750.15	8.57	12500			
Zr	2127.85	5.80	15030			
Hf	2506.15	12.00	10400			

€ from CRC Handbook(William, 2010)

Results and discussion

In the present work, in order to develop a statistical mechanically-based equation of state for refractory metals, some modifications are performed on the Tao-Mason EOS. This modification is important because it delivers two advantages: 1) these parameters are easier to measure than the critical parameters 2) the number of input parameters in the Tsonopolous' correlation (Tsonopoulos, 1974) are three (i.e., critical temperature, critical pressure, and acetric factor) while in the new correlation the number of input parameters reduce to two (i.e., T_m and ρ_m).

Besides we found that during examination of the TM EOS, k in Eq. (2) is a weak function of the acentric factor so that we can approximate k to 1.093. Because of this, A_2 was modified and approximated to a value of 1.64. Finally, the parameters α and b were correlated by means of Eqs. (12) and (13), respectively. In both equations, the input parameters, the melting temperature and the molar density in melting point, are more available than the Boyle temperature and volume. Generally, these modifications reduced the number of input parameters such as $B_2 \alpha$ and b from five (including critical temperature, critical pressure, acentric factor, Boyle temperature, and Boyle volume) to two (including melting temperature and the molar density in melting point).

Also, in this work we adjusted the parameter of λ for refractory metals by nonlinear regression method. This correlation presented as follow:

landa =

 $\frac{1}{a+b\times Tr+c\times Tr^2+d\times Tr^3+e\times Tr^4+f\times Tr^5}$

Where T_r is reduced temperature. The parameters that used for calculation of λ are scheduled in Table 1. Also the physical

(9)

Research article

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I [K]	P[Dar]	pcal [moi/Lit]	PExp [IIIOI/LII]			Dev% [wousazauen]
tantalum		aa a a	00.40	0.70	0.07	
3270	200	82.78	82.16	0.76	0.67	1.10
3350	200	81.25	81.43	0.22	1.06	1.52
3475	200	79.62	80.72	1.37	1.17	1.67
3675	200	78.56	80.02	1.83	0.84	1.39
3875	200	78.76	79.33	0.72	0.53	1.12
4075	200	78.64	78.65	0.01	0.26	0.86
4275	200	76.27	77.98	2.19	0.00	0.61
4525	200	76.85	77.33	0.62	0.50	0.09
4700	200	76 60	76.68	0.11	0.59	0.00
4900	200	76.09	76.05	0.06	0.79	0.24
5100	200	75 39	75.43	0.05	0.98	0.47
5250	200	74.81	74.81	0.00	0.00	0.47
5250	200	73.00	74.01	0.00	1.09	0.45
5450	200	73.99	79.02	0.29	0.00	0.07
5000	200	73.30	73.03	0.47	0.22	0.10
5800	200	72.50	72.46	0.14	0.39	0.09
6000	200	/1.//	71.89	0.17	0.55	0.33
6200	200	71.00	71.33	0.46	0.71	0.57
6400	200	70.25	70.24	0.01	0.11	0.05
6600	200	69.50	69.71	0.30	0.27	0.31
6800	200	68.73	68.67	0.09	0.32	0.17
7000	200	67.94	68.16	0.32	0.15	0.10
7200	200	67.12	67.17	0.08	0.72	0.34
7400	200	66.30	66.20	0.15	1.27	0.78
Total				0.45	0.61	0.57
rhenium	1					
3453	120	7 53	95.96	63	2 84	3 50
3500	120	7.46	95.87	1.66	2.64	3.20
3600	120	07.00	05.30	1.00	2.04	3.25
2700	120	97.09	95.50	1.00	2.04	3.20
3700	120	0.00	94.90	1.03	2.39	2.90
3800	120	95.86	94.50	1.44	2.31	2.85
3900	120	95.21	94.10	1.18	2.16	2.65
4000	120	94.52	93.63	0.95	2.10	2.54
4100	120	93.90	93.24	0.71	1.96	2.35
4200	120	93.34	92.70	0.69	1.99	2.33
4300	120	92.88	92.17	0.78	2.03	2.30
4400	120	92.50	91.64	0.94	2.07	2.28
4500	120	92.21	91.04	1.28	2.19	2.34
3453	200	97.53	100.35	2.81	1.59	1.01
3535	200	97.37	99.65	2.29	1.38	0.83
3699	200	96.56	98.95	.41	1.64	1.14
3840	200	5.59	97.83	2.29	1.32	0.88
4042	200	94 22	96.74	2.61	1.34	0.98
1216	200	93.25	95.27	12	0.77	0.00
4386	200	2.55	03.77	1 30	0.11	0.07
4500	200	2.33	02.62	0.50	0.03	0.07
4005	200	92.00	92.02	0.09	0.00	0.00
4000	200	1.75	91.79	0.04	0.10	0.24
4977	200	90.37	90.97	0.66	0.10	0.37
5180	200	0.54	90.39	0.17	0.46	0.91
5350	200	0.17	89.96	.23	0.81	1.41
5569	200	9.26	89.32	.06	1.14	1.95
5725	200	8.21	88.20	.01	0.62	1.59
Total				1.24	1.49	1.73
molybdenun	n					
2896	200	7.28	97.28	.00	1.10	0.99
3143	200	6.18	96.14	.04	0.48	0.46
3353	200		95.19	.12	0.00	0.03
3563	200	3 90	93 76	15	0.09	0.12
3773	200	2.63	92 70	07	0.16	0.09
3083	200	1 30	91 / 2	03	0.10	0.05
4102	200	0.10	00.19	.00	0.06	0.00
4193	200	0.10	90.10	.00	0.00	0.03
4403	200	9.03	00.90	.05	0.00	0.02
4613	200	7.88	87.87	.01	0.02	0.08
4823	200	6.63	86.66	.03	0.14	0.00
5033	200	5.13	85.13	.01	0.69	0.48
Total				0.05	0.26	0.21
				0.05	- Lesses	





Т [K]	ρ _{cal} [mol/Lit]	ρ _{Exp} [mol/Lit]	Dev% [TM]	Dev% [Eslami <i>et al.,</i> 2004]	Dev% [Mousazadeh, 2004]	
titanium						
1650	88.48	88.46	0.02	1.40	0.76	
1700	88.18	88.23	0.06	1.03	0.56	
1750	88.12	88.01	0.13	0.67	0.36	
1800	87.76	87.78	0.03	0.33	0.18	
1850	87.39	87.55	0.18	0.00	0.00	
1900	87.70	87.33	0.43	0.32	0.18	
1950	87.39	87.55	0.18	0.00	0.41	
2000	86.57	86.87	0.35	0.94	0.51	
2050	87.02	86.65	0.43	1.23	0.68	
Total			0.20	0.66	0.40	
Niobium						
232	84.94	84.95	0.01	0.99	0.78	
240	84.62	84.62	0.00	0.72	0.57	
250	84 40	84 40	0.00	0.57	0.46	
500	84.19	84.19	0.00	0.42	0.34	
2550	83.98	83.98	0.00	0.27	0.22	
2600	83 77	83 77	0.00	0.13	0.11	
2650	83.57	83 56	0.00	0.00	0.00	
2700	83.36	83 35	0.01	0.00	0.00	
2750	83.15	83.14	0.01	0.15	0.11	
2800	82.03	82.02	0.01	0.23	0.21	
2850	82 72	82.52	0.01	0.37	0.30	
2000	82.50	82.50	0.02	0.43	0.33	
2900	82.30	82.30	0.00	0.00	0.49	
Z950	02.20	02.29	0.01	0.71	0.30	
Total	n		0.01	0.43	0.55	
1950	60.00	60.00	0.00	2.45	2.20	
1000	00.00 69.75	69.74	0.00	2.40	2.29	
1900	69.50	69.50	0.01	2.11	2.03	
1950	69.44	69.44	0.00	1./0	1.70	
2000	00.44	00.44	0.01	1.40	1.04	
2000	00.29	00.29	0.00	1.10	1.01	
2100	00.15	00.14	0.01	0.00	1.00	
2150	08.00	08.00	0.01	0.00	0.00	
2200	07.00	07.85	0.00	0.28	0.03	
2250	07.70	67.70	0.1	0.00	0.41	
2300	67.55	67.55	0.00	0.27	0.20	
2350	07.40	b/.4U	0.00	0.53	0.00	
2400	07.25	07.20	0.02	0.79	0.22	
2450	0/.11	67.11	0.01	1.04	0.41	
2500	00.97	66.96	0.01	1.28	0.01	
2550	66.81	66.81	0.01	1.52	0.80	
2600	66.66	66.67	0.01	1./b	1.00	
2650	66.52	66.52	0.00	1.98	1.18	
2/00	66.39	66.37	0.02	2.21	1.3/	
2/50	66.21	66.22	0.01	2.42	1.54	
Iotal			0.01	1.29	1.01	
Hatnium		00.00				
2300	66.82	66.82	0.00	0.61	0.75	
2350	66.65	66.66	0.02	0.40	0.55	
2400	66.50	66.51	0.02	0.20	0.35	
2450	66.34	66.35	0.01	0.00	0.16	
2500	66.12	66.20	0.12	0.19	0.04	
2550	66.07	66.04	0.04	0.38	0.22	
2600	65.89	65.88	0.02	0.56	0.40	
2650	65.74	65.73	0.01	0.74	0.59	
Overall			0.03	0.39	0.38	

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$$\% Dev = \left(\frac{\rho_{exp} - \rho_{cal}}{\rho_{exp}}\right) \times 100$$

properties of all refractory metals are listed in Table 2. We performed the calculation of molar volume of all aforementioned refractory metals using the TM EOS. The result of the computed results has been gathered in Tables 3-4. It is obvious from these tables, that the agreement between the calculated density and the literature values (Jager *et al.*, 1992; Hixon & Winkler, 1992; Thevenin *et al.*, 1993; Ishikawa *et al.*, 2003) is quite suitable. In most cases, the obtained mean of the deviations for all refractory metals was found about 0.26%.

In order to evaluate the proposed correlation equation for the second virial coefficient, we have compared our results for these systems with those obtained using Eslami *et al.* (2004) and Mousazadeh (2007). The results of the calculations for all refractory metals are gathered in Tables 3-4. It is evident that, our results are in favor of the preference of the TM EOS over two other equation of state. The overall average absolute deviation that calculated by Eslami *et al.* (2004) and Mousazadeh (2007) are 0.73% and 0.66%, respectively.

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