



An Analytical Equation of State Extended to Copolymers

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Abstract

An analytical equation of state (EOS) entitled Ihm-Song-Mason (ISM) has been previously employed to calculate some thermodynamic properties of molten polymers including specific volume and isothermal compressibility (S.M. Hoseini, *Physical Chemistry & Electrochemistry*, 2 (2014) 56-65). This work extended that EOS to predict the volumetric properties of some molten ethylene copolymers including ethylene/1-octene, ethylene/1-butene (x_{ethylene} equal to 0.8543 and 0.563), ethylene/propene. The ability of the extended EOS was checked by comparing the results with 738 literature data points for the specific volumes over the temperature range from 422.35-505.89 K and pressure range from 0.1 up to 200 MPa. The average absolute deviation (AAD) of the calculated specific volumes from literature data was found to be 1.12%. The isothermal compressibility coefficients, κ_T have also been predicted using ISM EOS. From 180 data points examined, the AAD of estimated κ_T was equal to 8.48%. Our calculations on the volumetric and thermodynamic properties of studied copolymers reproduced the literature data with reasonably good accuracy.

Keywords: Equation of state, Copolymer melts, Thermodynamic properties

1. Introduction

The volumetric behaviour of any polymer yields the density (or its reciprocal, the specific volume (V)) as a function of pressure (P) and temperature (T). The knowledge of this behaviour is important as many phenomena of the behaviour of plastics depend on pressure and have associated changes in volume with them [1]. The PVT behaviour is of increasing importance since it is necessary

input for simulation software. Although some experimental data exist, prediction of thermophysical properties of polymers such as density and isothermal compressibility is still an important task considering they have been in focus as materials offering many highly promising applications. Under this circumstance, the development of equation of state (EOS) methods for the prediction of volumetric and thermodynamic properties of molten polymers can be highly useful.

Numerous efforts were made on the description of volumetric properties of molten

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polymers using EOS methods. An extensive survey of those efforts can be found in a book chapter written by Guerrieri et al.[2]. They reviewed a wide variety with EOSs from cubic [3-5] to those were based on the associating fluid theories (SAFT) [6].

Concerning molten copolymers, some authors [7, 8] have earlier assessed the performance of analytical EOSs for molten copolymers, where the dependent-parameters appearing in those EOSs were determined based on macroscopic (bulk) properties. However, the need for macroscopic scaling constants such as surface tension, melting temperatures and densities, for which their values are not easily accessible for most of polymers especially copolymers, limits the capability of those approaches.

This work is the successful extension of previous work [9] to molten copolymers for predicting their specific volumes and isothermal compressibilities. Earlier, an analytical EOS entitled Ihm-Song-Mason (ISM) [10] has been employed to predict some thermophysical properties of molten homopolymers. The distinctive feature of that work was applying the fixed free parameters, λ to ISM EOS [10]. That significance made it possible to reduce the number of input constants to be used in the EOS with respect to some similar published works related to this topic [11, 12]. In this paper, the performance of the ISM EOS based on the previous approach [9] is also checked by the estimation of isothermal compressibility coefficient of molten copolymers.

2. Theory

The general frame of the ISM EOS [7] can

be summarized as:

$$\frac{P}{\rho k_B T} = 1 + \frac{(B_2(T) - \alpha(T) \rho)}{1 + 0.2 \lambda b(T) \rho} + \frac{\alpha(T) \rho}{1 - \lambda b(T) \rho} \quad (1)$$

where, P is the pressure, ρ is the molar (number) density, $B_2(T)$ is the second virial coefficient, $\alpha(T)$ is the contribution of repulsive branch of pair potential function according to Weeks-Chandler-Anderson approach in perturbation theory of liquids [11]. This is based on the recognition that the structure of a liquid is determined primarily by repulsive forces, so that fluids of hard bodies can serve as useful reference states $b(T)$ reflects the van der Waals co-volume, $k_B T$ is the thermal energy per segment. λ is a free parameter which can be varied to get the high-density data of dense fluids at a singular point.

The ISM equation of state requires the usage of the second virial coefficient, $B_2(T)$, along with the parameters $\alpha(T)$ and $b(T)$. These parameters were previously evaluated in terms of microscopic scaling constants, i. e., pseudo-potential parameters including ε as the energy parameter and σ as the size parameter [9] as follows:

$$B_2(T) / (N_A \sigma^3) = F_2(k_B T / \varepsilon) \quad (2)$$

$$\alpha(T) / (N_A \sigma^3) = F_\alpha(k_B T / \varepsilon) \quad (3)$$

and

$$b(T) / (N_A \sigma^3) = F_b(k_B T / \varepsilon) \quad (4)$$

Where, F_2 , F_α and F_b are the empirical functions of reduced temperature ($T_r = k_B T / \varepsilon$).

In this work, those parameters were extended to copolymers by the help of pseudo-component parameters, ε^* and σ^* . It should be mentioned that the above mentioned temperature-dependent parameters, $B_2(T)$, $\alpha(T)$ and $b(T)$ reflect a measure of pair wise interactions operating between unlike hard-spheres per segment.

3. Results and discussion

3.1. Calculation of specific volumes

To utilize the proposed EOS two pseudo-component parameters of molten copolymers must be characterized. Table 1 comprises the pseudo-component parameters (σ^* , ε^*/k_B) of studied molten ethylene copolymers as well as their monomer molecular weight and mole fraction of ethylene, x_{ethylene} . Those parameters, (σ^* , ε^*/k_B) were optimized for each copolymer by fitting of isobaric specific volume data at 0.1 MPa, those were taken from literature [13] over the whole temperature range reported. The value of free parameter, λ is determined from experimental PVT data related to a datum point by the use of iterative method; the free parameter varies in order to get the following equality:

$$1 - \lambda b(T)\rho = \alpha(T)\rho \left[Z - 1 + \frac{(\alpha(T) - B_2(T)\rho)}{1 + 0.2 \lambda b(T)\rho} \right]^{-1} \quad (5)$$

We have taken the experimental data at high density like moderately high pressures

and low temperatures to convince the above-mentioned equality. Also, Table 1 demonstrates that, the free parameter is found to be depended on the chemical composition of copolymers of interest.

At first, we examined the performance of the ISM EOS for the prediction of specific volume of studied molten copolymers. For this purpose, the specific volume data of 4 molten copolymers were calculated and the results compared with the available literature data [13]. The results were reported in Tables 2-5 as the relative deviations (RD in %) of the predicted specific volumes from the literature data [13]. In Tables 2-5, bold-faces represent the PVT point at which the free parameter has been fixed.

Generally, our calculation results over the whole pressure-temperature range were summarized as the average absolute deviation (AAD in %) from literature data [13] in Table 6. As indicated by Table 6, from 738 data points examined for studied copolymers in a wide pressure range within 0.1-200 MPa and the temperature range from 423 to 505.89 K, AAD was found to be 1.12%.

Moreover, it has been found that the predicted specific volumes have been affected by pressure, systematically, as it's impressed from Tables 2-5. Since, the free parameter appearing in the ISM EOS has been fixed by a singular PVT point at high pressures; higher deviations have been appeared in the current

Table 1. Pseudo-component parameters of the studied molten copolymers to be used in Eqs. (2)-(4).

Copolymer	x_{ethylene}	MMW ^a (g/mol)	ε^*/k_B (K)	σ^* (nm)	λ
ethylene/1-octene	0.077	34.462	400.22	0.3633	0.7315
ethylene/1-butene	0.4046	34.878	405.14	0.3623	0.7350
ethylene/1-butene	0.1361	30.313	405.14	0.3623	0.6530
ethylene/propene	0.338	32.73	426.94	0.342	0.83329

^a MMW is the monomer molecular weight.

predictions at low and moderate pressures accordingly.

3.2. Calculation of thermodynamic properties

In this section, the relevant thermodynamic properties of molten copolymers to the first derivation of ISM EOS were also predicted. Typically, the isothermal compressibility

coefficient, κ_T is calculated using the isothermal pressure derivative of density/specific volume according to following equations:

$$\kappa_T = - \left(\frac{1}{V_S} \right) \left(\frac{\partial V_S}{\partial P} \right)_T = \frac{1}{\rho} \left(\frac{\partial \rho}{\partial P} \right)_T \quad (6)$$

We compared the calculated κ_T using the ISM EOS with those obtained from the Tait equation [14]. The results were reported

Table 2. Relative deviation (in %) ^a of the predicted specific volumes of ethylene/1-octene-copolymer with $x_{\text{Oct}} = 0.852$ from ISM EOS at several isotherms, compared with the literature data [13].

P/ MPa	423.1	433.01	443.28	453.42	463.8	473.36	483.63	493.64	503.31
0.1	-3.42	-2.98	-2.34	-1.62	-0.84	-0.01	0.98	2.11	3.30
10	-3.09	-2.65	-2.08	-1.45	-0.75	-0.05	0.83	1.77	2.75
20	-2.77	-2.35	-1.85	-1.27	-0.65	-0.05	0.74	1.53	2.35
30	-2.51	-2.13	-1.64	-1.16	-0.59	-0.07	0.63	1.33	2.01
40	-2.27	-1.92	-1.46	-1.01	-0.54	-0.06	0.55	1.18	1.76
50	-2.06	-1.74	-1.31	-0.91	-0.44	-0.06	0.51	1.05	1.57
60	-1.85	-1.56	-1.17	-0.80	-0.39	-0.02	0.47	0.96	1.41
70	-1.65	-1.40	-1.03	-0.70	-0.33	0.00	0.46	0.86	1.28
80	-1.47	-1.24	-0.91	-0.59	-0.27	0.02	0.43	0.84	1.17
90	-1.31	-1.09	-0.78	-0.51	-0.21	0.05	0.42	0.79	1.10
100	-1.15	-0.96	-0.67	-0.42	-0.14	0.08	0.43	0.75	1.03
110	-0.99	-0.82	-0.55	-0.32	-0.08	0.13	0.43	0.72	0.98
120	-0.82	-0.69	-0.43	-0.22	-0.01	0.18	0.46	0.72	0.94
130	-0.68	-0.54	-0.32	-0.13	0.07	0.23	0.49	0.72	0.92
140	-0.52	-0.41	-0.20	-0.03	0.15	0.30	0.52	0.73	0.90
150	-0.37	-0.29	-0.08	0.07	0.23	0.35	0.56	0.74	0.90
160	-0.22	-0.15	0.03	0.18	0.32	0.42	0.60	0.77	0.91
170	-0.08	-0.03	0.15	0.28	0.40	0.48	0.64	0.81	0.91
180	0.04	0.11	0.26	0.37	0.48	0.55	0.69	0.82	0.93
200	0.00^b	0.34	0.46	0.56	0.63	0.68	0.78	0.90	0.96

$$^a R D = \left(\frac{V^{Calc.} - V^{Exp.}}{V^{Exp.}} \right) 100$$

^b Bold face is the PVT point, at which λ has been fitted

Table 3. Relative deviation (in %)^a of the predicted specific volumes of ethylene/1-butene-copolymer with $x_{1\text{-butene}} = 0.864$ using the proposed EOS at several isotherms, compared with the literature data [13].

<i>P</i> / MPa	425.24	434.75	444.75	455.35	465.22	475.34	485.06	495.15	505.89
0.1	-5.14	-4.78	-4.35	-3.89	-3.30	-2.57	-1.91	-1.09	-0.10
10	-4.62	-4.25	-3.83	-3.36	-2.81	-2.12	-1.50	-0.72	0.18
20	-4.15	-3.78	-3.37	-2.87	-2.38	-1.73	-1.15	-0.41	0.43
30	-3.74	-3.38	-3.00	-2.52	-2.01	-1.44	-0.88	-0.21	0.57
40	-3.38	-3.03	-2.65	-2.21	-1.73	-1.16	-0.66	-0.03	0.69
50	-3.05	-2.71	-2.34	-1.92	-1.47	-0.93	-0.43	0.12	0.79
60	-2.74	-2.41	-2.06	-1.67	-1.23	-0.73	-0.26	0.25	0.88
70	-2.46	-2.14	-1.81	-1.43	-1.03	-0.55	-0.12	0.40	0.95
80	-2.19	-1.89	-1.58	-1.21	-0.83	-0.38	0.02	0.50	1.06
90	-1.95	-1.66	-1.36	-1.01	-0.64	-0.22	0.16	0.61	1.12
100	-1.71	-1.43	-1.16	-0.82	-0.47	-0.08	0.28	0.70	1.18
110	-1.49	-1.22	-0.96	-0.64	-0.32	0.06	0.41	0.78	1.24
120	-1.28	-1.01	-0.76	-0.47	-0.16	0.21	0.51	0.88	1.30
130	-1.08	-0.82	-0.60	-0.31	0.00	0.33	0.63	0.96	1.37
140	-0.88	-0.63	-0.42	-0.14	0.14	0.46	0.72	1.05	1.43
150	-0.69	-0.45	-0.25	0.00	0.29	0.59	0.84	1.14	1.49
160	-0.51	-0.27	-0.09	0.16	0.42	0.71	0.93	1.22	1.55
170	-0.34	-0.11	0.07	0.30	0.55	0.81	1.04	1.31	1.63
180	-0.16	0.07	0.22	0.44	0.67	0.93	1.14	1.38	1.69
190	0.00^b	0.21	0.36	0.58	0.80	1.04	1.24	1.47	1.76
200	0.15	0.38	0.50	0.70	0.91	1.15	1.33	1.54	1.83

$${}^a R D = \left(\frac{V^{Calc.} - V^{Exp.}}{V^{Exp.}} \right) 100$$

^b Bold face is the PVT point, at which λ has been fitted

Table 4. Relative deviation (in %)^a of the predicted specific volumes of ethylene/1-butene-copolymer with $x_{1\text{-butene}} = 0.595$ using the proposed EOS at several isotherms, compared with the literature data [13].

<i>P</i> / MPa	424.48	433.74	443.82	454.18	464.52	474.15	484.04	493.79	504.43
0.1	-3.04	-2.45	-1.67	-0.78	0.22	1.18	2.40	3.79	5.53
10	-2.74	-2.18	-1.46	-0.67	0.23	1.10	2.15	3.30	4.71
20	-2.47	-1.94	-1.28	-0.56	0.26	1.04	1.99	2.94	4.13
30	-2.24	-1.76	-1.14	-0.49	0.26	0.96	1.79	2.64	3.65
40	-2.04	-1.59	-1.02	-0.43	0.26	0.89	1.63	2.38	3.30
50	-1.87	-1.44	-0.91	-0.37	0.25	0.82	1.50	2.17	2.96
60	-1.70	-1.31	-0.82	-0.31	0.25	0.77	1.38	1.98	2.69
70	-1.55	-1.18	-0.72	-0.26	0.26	0.73	1.27	1.83	2.47
80	-1.40	-1.06	-0.63	-0.20	0.26	0.69	1.20	1.69	2.28
90	-1.27	-0.95	-0.56	-0.15	0.27	0.67	1.13	1.58	2.12
100	-1.13	-0.83	-0.47	-0.09	0.30	0.66	1.09	1.49	1.99
110	-1.01	-0.73	-0.39	-0.03	0.32	0.66	1.04	1.40	1.87
120	-0.89	-0.62	-0.30	0.01	0.34	0.66	1.01	1.35	1.77
130	-0.77	-0.51	-0.23	0.07	0.38	0.66	0.99	1.30	1.69
140	-0.65	-0.41	-0.15	0.12	0.42	0.68	0.97	1.26	1.62
150	-0.53	-0.31	-0.07	0.19	0.45	0.69	0.97	1.23	1.56
160	-0.42	-0.21	0.03	0.25	0.50	0.71	0.97	1.21	1.52
170	-0.31	-0.12	0.10	0.32	0.54	0.73	0.98	1.19	1.49
180	-0.21	-0.01	0.19	0.38	0.59	0.77	0.98	1.18	1.46
200	0.00^b	0.17	0.34	0.50	0.68	0.82	1.01	1.18	1.42

$${}^a R D = \left(\frac{V^{Calc.} - V^{Exp.}}{V^{Exp.}} \right) 100$$

^b Bold face is the PVT point, at which λ has been fitted.

Table 5. Relative deviation (in %)^a of the predicted specific volumes of ethylene/propene-copolymer with $x_{\text{propene}} = 0.662$ using the proposed EOS at several isotherms, compared with the literature data [13].

<i>P</i> / MPa	422.35	432.5	442.35	452.05	462.27	472.37	482.18	492.51	502.28
0.1	-3.24	-2.55	-1.84	-0.96	0.11	1.29	2.59	4.16	5.89
10	-2.92	-2.26	-1.61	-0.80	0.14	1.16	2.27	3.59	5.00
20	-2.63	-2.01	-1.38	-0.67	0.17	1.06	2.04	3.18	4.36
30	-2.39	-1.81	-1.24	-0.56	0.16	0.96	1.82	2.82	3.84
40	-2.18	-1.64	-1.12	-0.49	0.20	0.89	1.64	2.52	3.42
50	-1.99	-1.47	-1.01	-0.42	0.20	0.86	1.50	2.29	3.08
60	-1.82	-1.32	-0.91	-0.35	0.21	0.81	1.43	2.09	2.79
70	-1.66	-1.20	-0.81	-0.30	0.22	0.76	1.32	1.96	2.54
80	-1.51	-1.07	-0.72	-0.24	0.24	0.74	1.24	1.82	2.39
90	-1.37	-0.95	-0.62	-0.19	0.26	0.72	1.17	1.71	2.22
100	-1.22	-0.84	-0.53	-0.13	0.28	0.71	1.12	1.61	2.08
110	-1.10	-0.72	-0.44	-0.07	0.31	0.70	1.09	1.53	1.96
120	-0.96	-0.61	-0.34	-0.02	0.35	0.71	1.06	1.48	1.86
130	-0.83	-0.51	-0.26	0.04	0.38	0.71	1.03	1.43	1.78
140	-0.70	-0.40	-0.16	0.11	0.43	0.73	1.03	1.38	1.71
150	-0.58	-0.28	-0.06	0.17	0.47	0.75	1.01	1.36	1.66
160	-0.47	-0.18	0.03	0.25	0.51	0.76	1.03	1.32	1.62
170	-0.34	-0.07	0.12	0.31	0.56	0.79	1.04	1.30	1.57
180	-0.22	0.03	0.21	0.39	0.61	0.82	1.05	1.30	1.54
190	-0.11	0.12	0.30	0.45	0.67	0.85	1.06	1.29	1.52
200	0.00^b	0.22	0.38	0.51	0.72	0.88	1.08	1.27	1.51

$${}^a R D = \left(\frac{V^{Calc.} - V^{Exp.}}{V^{Exp.}} \right) \cdot 100$$

^b Bold face is the PVT point, at which λ has been fitted

Table 6. The AAD (in %) of the predicted specific volumes of the entire molten copolymers studied in this work, using the ISM EOS, compared with the literature data [13].

Polymer	ΔP (MPa)	ΔT (K)	NP ^a	AAD (%)
ethylene/1-octene	0.1-200	423-503	180	1.050
ethylene/1-butene ^c	0.1-200	425- 505.89	189	1.18
ethylene/1-butene ^d	0.1-200	424.5- 504.43	180	1.12
ethylene/propene	0.1-200	422.35 -502.3	189	1.14
Overall			738	1.12

^a NP represents the number of data points examined

$$^b \text{AAD} = 100/\text{NP} \sum_{i=1}^{\text{NP}} |V_{i \text{ Cal.}} - V_{i \text{ Exp.}}| / V_{i \text{ Exp.}}$$

$$^c x_{\text{ethylene}} = 0.13607$$

$$^d x_{\text{ethylene}} = 0.4046$$

in Table 7 as the relative deviations (RD in %) of the predicted κ_T of ethylene/1-butene copolymer ($x_{\text{ethylene}} = 0.4046$) from those derived by the Tait equation [16]. From 180 data points examined, the AAD of estimated κ_T was equal to 8.48%. According to Table 7, the maximum value of relative deviations (RD) in predicting the isothermal compressibility coefficients was found to be 24.95%. Generally, the reason for such large deviations is somewhat due to some systematic errors appearing in parameter determinations. As a matter of fact, pseudo-component parameters of studied copolymers were fitted against the density/volume data rather than density/volume derivative of the EOS of interest, i.e., κ_T is closely related to the isothermal density/volume derivative of the EOS, the large deviations from the results of Tait equation are reasonably expectable.

In this paper, the performance of the ISM based on the previous approach [9] has been checked by the estimation of specific volumes and isothermal compressibility coefficients

of molten copolymers with reasonably good AAD.

4. Conclusion

Finally, an analytical EOS has been extended to predict the volumetric and thermodynamic data of molten copolymers. Knowing only two molecular scaling constants being the pseudo-component parameters, ε^*/k_B and σ^* , is sufficient to determine the temperature-dependent parameters of the ISM EOS to be employed for molten copolymers. Our calculations on the volumetric and thermodynamic properties of studied copolymers reproduced the literature data over the whole liquid range with reasonably good accuracy.

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Table 7. Relative deviation (in %)^a of the predicted isothermal compressibility coefficients (κ_T) of ethylene/1-butene copolymer ($x_{\text{ethylene}} = 0.4046$) using the ISM EOS at several isotherms, compared with the those obtained from the Tait equation [14].

<i>P</i> / MPa	425.24	434.75	444.75	455.35	465.22	475.34	485.06	495.15	505.89
0.1	-24.44	-19.96	-14.56	-8.31	-1.17	6.54	15.86	24.95^b	22.13
10	-23.21	-18.79	-13.54	-7.58	-0.92	6.06	14.22	23.52	20.52
20	-22.21	-17.87	-12.77	-7.06	-0.81	5.60	12.92	21.01	20.06
30	-21.40	-17.14	-12.19	-6.73	-0.82	5.14	11.81	19.03	19.77
40	-20.75	-16.58	-11.77	-6.52	-0.92	4.67	10.84	17.39	19.19
50	-20.22	-16.14	-11.48	-6.42	-1.08	4.20	9.94	15.98	18.65
60	-19.80	-15.81	-11.27	-6.40	-1.28	3.71	9.11	14.73	18.23
70	-19.46	-15.56	-11.15	-6.44	-1.53	3.22	8.33	13.59	17.62
80	-19.20	-15.38	-11.10	-6.53	-1.82	2.73	7.58	12.54	17.18
90	-19.00	-15.27	-11.09	-6.67	-2.12	2.24	6.85	11.56	16.87
100	-18.86	-15.20	-11.13	-6.84	-2.45	1.74	6.16	10.64	15.67
110	-18.75	-15.18	-11.21	-7.04	-2.79	1.25	5.48	9.76	14.54
120	-18.69	-15.19	-11.32	-7.27	-3.15	0.75	4.83	8.93	13.49
130	-18.67	-15.24	-11.45	-7.51	-3.51	0.26	4.19	8.12	12.50
140	-18.67	-15.31	-11.61	-7.77	-3.89	-0.23	3.57	7.36	11.55
150	-18.70	-15.40	-11.79	-8.04	-4.26	-0.72	2.96	6.61	10.65
160	-18.75	-15.52	-11.98	-8.32	-4.64	-1.20	2.36	5.89	9.79
170	-18.82	-15.65	-12.19	-8.61	-5.03	-1.68	1.78	5.20	8.96
180	-18.91	-15.80	-12.40	-8.91	-5.41	-2.15	1.21	4.52	8.17
200	-19.13	-16.13	-12.87	-9.52	-6.19	-3.09	0.09	3.23	6.65

$${}^a RD = \left(\frac{\kappa_T^{Calc.} - \kappa_T^{Tait}}{\kappa_T^{Tait}} \right) \cdot 100$$

^b Bold face is the maximum %RD in the predicted κ_T .

Nomenclature and units

List of symbols

AAD	average absolute deviation (%)
k_B	Boltzmann's constant ($J K^{-1}$)
NP	number of data points
P	pressure (Pa)
T	Absolute temperature (K)
V	Specific molar volume ($m^3 mol^{-1}$)
MMW	monomer molecular weight ($g \cdot mol^{-1}$)
R	Universal gas constant ($J mol^{-1} \cdot K^{-1}$)
B_2	second virial coefficient/ (m^3)
b	co-volume/ (m^3)
α	repulsive contribution to pair potential function/ (m^3)
N_A	Avogadro's number

Greek letters

ρ	molar density ($mol m^{-3}$)
σ	segment diameter (nm)
ε	dispersive energy parameter (J)
σ^*	pseudo-segment diameter (nm)
ε^*	pseudo-dispersive energy parameter (J)
λ	free parameter

Superscripts

Calc	Calculated values
Exp	Experimental value

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