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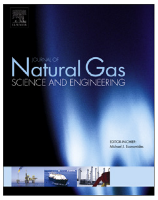

Prediction model of elemental sulfur solubility in sour gas mixtures

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ABSTRACT

Predicting the solubility of elemental sulfur in sour gas mixtures is one of the most important issues in sour gas reservoirs development. Many experiments have shown the relationship between sulfur solubility and sour gas properties at different temperatures and pressures. However, the solubility model that can be used to predict sulfur solubility at different reservoir temperatures and pressures is rare. Nowadays, Roberts's solubility formula, according to fitting two groups of experiment data, is often used in sour gas reservoirs. Thus, building a solubility formula that contains more experimental data and experimental conditions is very critical. In this paper, based on the correlation formulas for the solubility of various solutes in super critical solvents, a new sulfur solubility formula is built by using a large number of experimental data. Through the comparison and analysis for the new solubility formula and Roberts's solubility formula, the calculation results utilizing the new solubility formula are closer to the experimental data. Therefore, the new formula can be used to predict sulfur solubility in sour gas reservoirs at different temperatures and pressures accurately. The new solubility model can calculate the change of sulfur solubility in sour gas well production and help reservoir engineers to develop the plan of sour gas reservoir development.

1. Introduction

Elemental sulfur precipitates from sour gas with reservoir pressure and temperature decreasing. Sulfur deposition in the formation may significantly reduce the inflow performance of sour gas wells. Sulfur solubility in sour gas mixtures is one of the most important problems in sour gas reservoirs development. The sulfur solubility data in sour gas mixtures at different temperatures and pressures can be accurately obtained by experiments, however, experiments methods needs large amounts of time and devices, what's more, sour gas mixtures used in the experiment are toxic. If a theoretical formula can be built to predict sulfur solubility in sour gas well production, it will help reservoir engineers develop sour gas reservoirs better.

By analyzing experimental data of Brunner and Woll, Roberts (1997) built a theoretical formula of sulfur solubility on the basis of Chrastil's solubility model (Chrastil, 1982). As it is convenient and practical, this solubility model is widely used in sour gas reservoir development. However, the sulfur solubility model is acquired from only two groups of experiments data and the application conditions of this model are not given. Thus, in this paper, based on the principles of Chrastil solubility model, using amount of experimental data under different experimental conditions, a scientific mathematical method is used to build a new sulfur solubility model. The new solubility model can be used to describe the change of sulfur solubility in different components of sour gas mixtures under different temperatures and pressures.

2. Analysis of Chrastil solubility model and Roberts's empirical formula

Based on associative law and entropy principle, an empirical solubility equation for predicting the solubility of solid and liquid in a high-density gas was presented (Chrastil, 1982). It can be used to evaluate the relationships between solubility and pressure (temperature). As the theory model shows:

$$C_r = \rho^k \exp(a/T + b) \tag{1}$$

The density of sour gas mixtures can be expressed as follows:

$$\rho = M_a \gamma_{gp}/(ZRT) = 3.484 \times 10^3 \times \left(\gamma_{gp}\right)\Big/(ZT) \tag{2}$$

Where: C_r-sulfur solubility in sour gas mixtures, g/L; ρ −gas density, kg/m^3;T − reservoir temperature, K; k − empirical

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constant; M_a – molecular weight of dry air, 28.97; γ_g – gas relative density; Z – gas deviation factor (average value); R – gas constant, 0.008315 MPa m^3/kmol.k; P – reservoir pressure, MPa.

Based on the principle of Chrastil solubility model, using Brunner and Woll (1980), Brunner et al. (1988) experimental data (Table 1), an empirical equation of sulfur solubility in sour gas mixtures is obtained by Roberts in 1997.

$$C_r = \rho^4 \exp(-4666/T - 4.5711) \tag{3}$$

Due to the conveniences and practicability of Roberts's solubility model, the model is widely used in sour gas reservoir development. However, the solubility model from only two groups of experiments. In fact, experiment data of the sulfur solubility used by Roberts is sulfur dissolution in hydrogen sulfide but not sulfur dissolution in sour gas mixtures. Thus, sulfur solubility model in sour gas mixtures should be carefully deduced and calculated.

3. Experimental data of sulfur solubility

Sulfur solubility experiments under different experimental conditions have been completed by different scholars. (Brunner and Woll, 1980; Brunner et al., 1988; Kennedy and Wieland, 1960; Roof, 1971; Swift and Manning, 1976; Zeng et al., 2005; Gu et al., 1993; Yang et al., 2009; Sun and Chen, 2003). Experiments data of sulfur solubility is shown and listed in Table 2.

When using Chrastil solubility model, in order to make the correlation coefficient fit the conditions of reservoir better, some basic principles are built to choose experimental data.

(1) The integrity of the experimental system;
(2) Experimental temperatures and pressures are close to reservoir conditions;
(3) Gas density should be calculated or measured accurately in experimental system.

By comparing and analyzing the experimental data, list 1 and 2 in Table 2 are chosen to achieve solubility empirical formula. The other experimental data, which cannot be chosen in Table 2, the reason is that they are close to list 1 and list 2 or out of the range of reservoir conditions. A new sulfur solubility model is established and large amounts of experimental data are used to verify its accuracy.

4. Fitting method of Chrastil solubility model

The Chrastil solubility model contains coefficient "k", "a" and "b". In this paper, a new sulfur solubility model is established and the mathematics method of data-fitting is as follows:

Taking the logarithms of each side of Formula (1):

$$\ln C_r = k\ln\rho + (a/T + b) \tag{4}$$

Formula (4) can be simply changed into:

$$\ln\rho = \ln C_r/k - (a/T + b)/k \tag{5}$$

Thus, at a certain temperature, a log–log plot of the solubility versus the density should yield a straight line with a slope of 1/k and an intercept of $-(a/T + b)/k$.

Table 1
Experimental date in Roberts sulfur solubility model.

Experimental system	Gas components
BW1	66% CH_4, 20%H_2S, 10% CO_2, 4%N_2
BW3	81% CH_4, 6%H_2S, 9% CO_2, 4%N_2

Table 2
Experimental data of sulfur solubility.

List	Author	Temperature (°C)	Pressure (MPa)	Gas components
1	Brunner and Woll (1980)	100–160	10–60	H_2S and CH_4, CO_2, N_2 at different ratio of mixed gas
2	Brunner et al (1988)	125–212	6. 7–155	H_2S, CH_4, CO_2 and N_2 at different ratio of mixed gas
3	Kennedy&W ireland	93.3–121.1	7–41. 4	Pure CH_4, CO_2, H_2S, CH_4 and different ratio of the four mixed gas
4	Roof	43.3–110	7–31	Pure H_2S
5	Swift et a.l	121–204	34. 5–138	Pure H_2S
6	P. Zeng	80–160	10–60	H_2S, CH_4, CO_2 and N_2 at different ratio of mixed gas
7	M.-X. Gu	90–110	10–50	H_2S, CH_4, CO_2 and N_2 at different ratio of mixed gas
9	X.-F. Yang	100	16–36	H_2S, CH_4, CO_2 and N_2 at different ratio of mixed gas
8	C.-Y. Sun	30–90	20–45	CH_4, H_2S, CO_2

Due to the coefficient "a" and "b" have no relationship with T, at constant temperatures T_1 and T_2:

$$-(a/T_1 + b)/k = m_1 \tag{6}$$

$$-(a/T_2 + b)/k = m_2 \tag{7}$$

The coefficient "a" and "b" can be obtained by simply calculation:

$$a = kT_1T_2(m_1 - m_2)/(T_1 - T_2) \tag{8}$$

$$b = k(m_2T_2 - m_1T_1)/(T_1 - T_2) \tag{9}$$

5. Establishing of sulfur solubility model in sour gas mixtures

According to the temperatures and pressures in actual sour gas reservoirs, experimental data (list 1 and 2) in Table 2 are chosen to establish the sulfur solubility model. In order to prove the correctness of the new solubility model, amount of experimental data are used to be calculated and compared. Hollow symbol is a representative of new solubility calculation values and the solid symbol is a representative of experimental data in Figs. 2, 4 and 6 and 8.

5.1. *The sulfur solubility in sour gas mixtures (1)*

Gas compositions (in volume): 66%CH_4, 20%H_2S, 10%CO_2, 4%N_2. The gas density is 1.028 kg/m^3 (101.325 kPa, 273.15K).

Based on the above theories and principles, a new sulfur solubility model is established and the coefficients are obtained from Tables 3 and 4.

With the increasing of gas density, the sulfur solubility increases in Figs. 1 and 2. However, when the gas density is up to 200 kg/m^3, an inflection point appears. The new solubility model can fit the

Table 3
Correlation coefficients in solubility model.

Density/ρ	k	a	b
<200 (kg/m^3)	1.592044	–2737.23	–8.89768
>200 (kg/m^3)	3.288695	–4880.74	–12.4969