

Viscosity Models and Effects of Dissolved CO₂

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ABSTRACT

A comprehensive study is carried out on viscosity modeling for the geologic sequestration of CO₂ in the pressure and temperature range 1-600 bar, and 20°-105° C, respectively. For the liquid phase we present viscosity models for pure water (H₂O), brine (H₂O+NaCl), H₂O+CO₂, H₂O+NaCl+CO₂, and typical sea water compositions. In each case, we attempted to develop very accurate formulations, but having fewer parameters than existing models for more efficient computation. Effects of dissolved CO₂ are studied extensively. We have found that deviations can be 2-8% in liquid phase viscosity calculations if CO₂ dissolution is neglected. In addition, a vapor phase model is also suggested.

1. Introduction

Increasing atmospheric concentrations of greenhouse gases are suspected of causing a gradual warming of the Earth's surface and potentially disastrous changes to global climate. Because CO₂ is a major greenhouse gas, storing it in subsurface formations is being explored as a viable option to limit the accumulation of greenhouse gases in the atmosphere. Carbon sequestration, sometimes broadly referred as carbon management, is a way to reduce greenhouse gas emissions while still enjoying the benefits of fossil fuel use. This is a complementary approach to the current CO₂ mitigation efforts of improved energy efficiency and increased use of non-carbon energy sources. These days, much attention is given in the carbon management option because it is very compatible with the large energy production and delivery infrastructure now in place, and because non-fossil energy sources face large barriers; renewables are very expensive and nuclear has safety concerns. Sequestration covers technologies that capture carbon at its source (e.g., power plants, industrial processes) and directs it to non-atmospheric sinks (e.g., depleted oil and gas reservoirs, deep

saline formations, coal seams, hard rock caverns, deep ocean), as well as processes that increase the removal of carbon from the atmosphere by natural processes (e.g., forestation) being widely available, and currently having a lack of effective uses due to the great potential of storage. The most promising places for sequestration are aquifers (Dirik et al., 2004; Herzog, April, 2001) Accurate evaluation of the capacity of a saline aquifer for CO₂ sequestration and the fate of the injected fluids in sedimentary basins require analysis of the thermo-physical properties of CO₂ and brine. The thermo-physical properties include thermodynamic properties, e.g., PVT (pressure-volume-temperature) behaviors; and transport properties, e.g., viscosities, thermal conductivities, and diffusion coefficients. Our previous studies (Islam and Carlson, 2012a, b, c) contain comprehensive investigations on PVT behaviors. This study focuses on one of primary transport properties, viscosity, and presents some simpler and more efficient tools to compute the viscosity of aqueous and gaseous phases in CO₂ sequestration.

For any multiphase flow system viscosity plays an important role. Viscosity characterizes the fluids' resistance with respect to deformation under shear stress (Class, 2007). The lower a fluid's viscosity, the lower its resistance to flow and displaces one fluid by another. If the viscosity of the injected CO₂ is higher than the viscosity of the CO₂ and brine that is in the aquifer, the displacement front is stable. On the other hand if the viscosity of the injected CO₂ is lower, the displacement front can become unstable. Therefore, accurate prediction of viscosity is extremely important. Though not addressing simulation of CO₂ sequestration, Davani et al., (2009) and Hernandez et al. (2002) have comprehensively investigated the sensitivity of reservoir simulations to uncertainties in viscosity for both liquid and vapor phases.

In general, the viscosity change of brine with CO₂ saturation is neglected in the simulators developed (Hassanzadeh et al., 2008; Lu and Lichtner, 2007; Ozgur and Gumrah, 2010; Piri et al., 2005; Pruess, 2005). This is because there is no model available in the literature for the effect of dissolved CO₂ on water/brine viscosity. However, in reality viscosity is directly related to density, and dissolution of CO₂ may cause density variations of 2-3% (Islam and Carlson, 2012c; Mao and Duan, 2009). When injecting CO₂, the plume has a tendency to flow upward. However, a small amount of

CO₂ will dissolve into the water; under the influence of the small density difference, that water has the tendency to flow downward (Fleury and Deschamps, 2008). (Tumasjan et al., 1969) showed that the viscosity of water varies from 1.0 to 1.3 centipoise for 4% (by weight) dissolved CO₂. Thus it is important that CO₂ dissolution be considered while modeling viscosity. In this paper we will present some simple empirical formulations for computing the viscosity of pure water, brine (H₂O+NaCl and H₂O+NaCl+CO₂), and typical sea water (having 3.5% salinity) for the pressure and temperature range of a saline aquifer at 1-600 bar, 20°-105° C, respectively (Spycher et al., 2003). In addition, we will analyze how viscosity varies quantitatively for CO₂ dissolution in the aqueous phase, and will also recommend viscosity calculation for the gaseous phase.

2. Viscosity of Pure Water

Very well established formulations for the viscosity of H₂O are available for wide pressure and temperature range (ASME 1967; 2003; Hendricks et al., 1977; Kestin, 1980; Kestin and Whitelaw, 1966; Nagashima, 1977; Sengers and Watson, 1986). The most recent is the IAPWS Formulation 2009 (Huber et al., 2009), which covers temperature and pressure up to 900° C, and 10000 bar, respectively. Here, simpler correlation with fewer parameters for the viscosity calculation applicable in relevant P - T range is proposed. The correlation is given by function of P and T , where

$$\mu_{H_2O} = a_0 + \sum_{i=1}^3 b_i \exp(-c_i T) + P \sum_{i=0}^3 d_i (T - 293.15)^i \quad (1)$$

Parameters of Eq. 1 are estimated by regressing generated data from the IAPWS Formulation 2009 (IAPWS09). The coefficients a , b , c , and d are reported in Table 1. P is in MPa. Viscosity data of pure water can be regenerated by Eq. 1 with a maximum 0.05% deviation as compared

to IAPWS09, where deviation = $\frac{lit - cal}{lit} \times 100$. Figure

1(a-d) show comparisons of data at different isotherms (20°, 50°, 80°, and 100° C). From this figure it is clear that maximum divergence (~0.05%) occurs at 100° C at low pressures (<5 bar). Otherwise divergences lie within 0.04%.

Table 1. Coefficients of Eq. 1.

i	a	b	c	d
0	9.03591045e+01			-1.22757462e-01
1		3.40285740e+04	1.40090092e-02	2.15995021e-02
2		8.23556123e+08	4.86126399e-02	-3.65253919e-04
3		-9.28022905e+08	5.26696663e-02	1.97270835e-06

3. Viscosity of H₂O+NaCl

Recently Mao and Duan (2009) have done very nice viscosity modeling work for brine water (H₂O+NaCl). Their model covers the P - T range up to 1000 bar and 350° C, and ionic strength

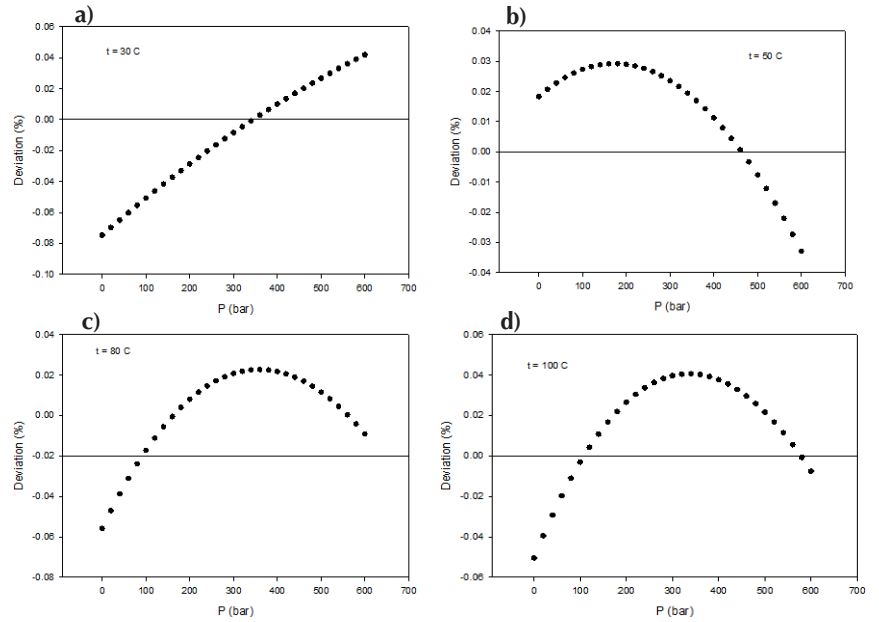


Figure 1 (a-d). Deviation between calculated data from IAPWS09 and Eq. 1.

of up to 6.0 molality. Their formula can reproduce the literature values within 1% deviation. However, in their model they have used IAPWS97 (Wagner et al., 2000) for density calculation of water. Instead, to reduce the number of parameters for efficient computation of water density, we recommend following equation,

$$\rho_{H_2O} = a_0 + \sum_{i=1}^3 b_i 10^{c_i T} + \sum_{i=1}^2 d_i P^i \quad (2)$$

Parameters of Eq. 2 are predicted by regressing generated data from IAPWS97, with this modification of Mao and Duan's (2009) formulation we reproduce their values with maximum 0.1% deviation. P is in MPa. Table 2 shows the parameter's values. Figure

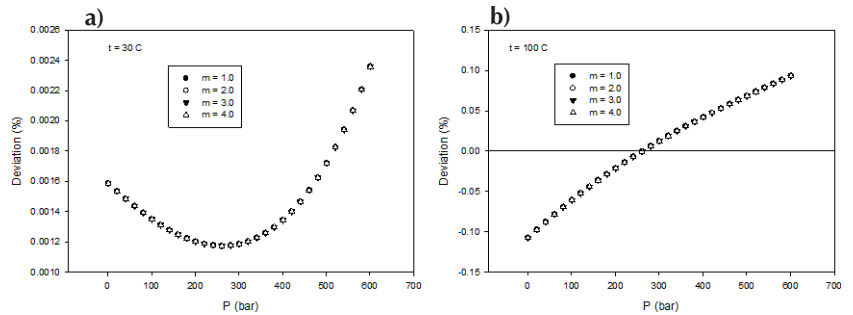


Figure 2 (a,b). Deviation between calculated data from Mao and Duan (2009), and Eq. 2.

2(a,b) show these comparisons for two different temperatures, 30° and 100° C, and different molalities of NaCl. From this figure we can also observe that deviation increases (~0.1%) at higher temperatures; and there is no variation due to changing molality.

Table 2. Coefficients of Eq. 2.

i	a	b	c	d
0	1.34136579e+02			
1		-4.07743800e+03	-5.56126409e-03	4.45861703e-01
2		1.63192756e+04	-1.07149234e-02	-4.51029739e-04
3		1.37091355e+03	-5.46294495e-04	

4. Viscosity of H₂O+CO₂

Kumagai et al. (1998) measured the viscosity of water containing up to 4.8% (by weight) CO₂ at pressures up to 400 bar and the temperatures from 0°-50° C. They presented their experimental results only in graphical form, and therefore, although there is chance of having uncertainty in picking exact numeric value from graphs, we did so for our modeling purposes, yielding the following equation:

$$\mu_r = 1 + \frac{\sum_{i=1}^2 a_i x_{CO_2}^i}{\sum_{i=0}^1 b_i T^i} \quad (3)$$

where

$$\mu_{H_2O+CO_2} = \mu_r \times \mu_{H_2O} \quad (4)$$

The parameters are reported in Table 3.

Table 3. Coefficients of Eq. 3.

i	a	b
0	7.632609119e+02	-1.047187396332e+04
1	-9.46077673946e+03	3.6.8325597e+01

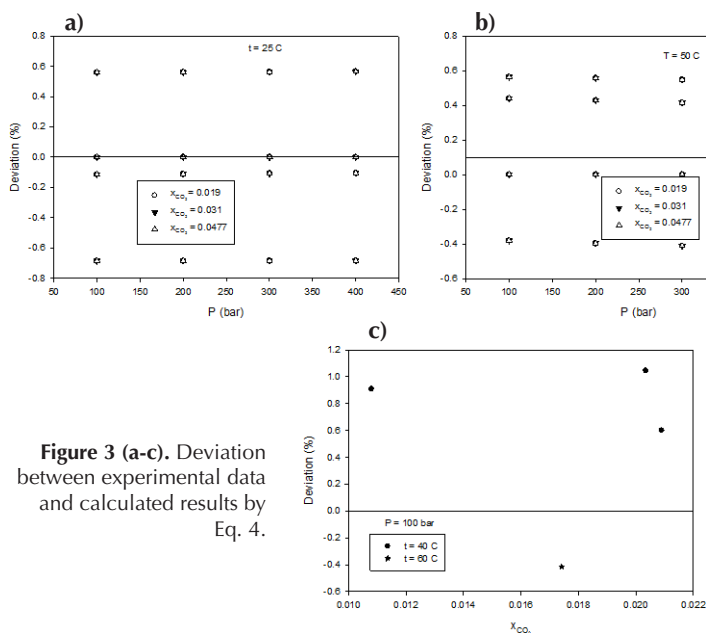


Figure 3 (a-c). Deviation between experimental data and calculated results by Eq. 4.

Direct experimental values from Bando et al. (2004) were also used for the data reduction process. The error of calculated data from this model is shown in Figure 3(a,b); the maximum deviation reported is 0.68% compared to the data from Kumagai et al. (1998). Comparative results of Figure 3c were produced with respect to the data by Bando et al. (2004). The author's study clearly reveals that, for temperatures greater than 25° C, the effect of pressure on the viscosity of water with dissolved CO₂ is nil and, as the temperature increases (>50° C), the effect of dissolution of CO₂ becomes less important.

5. Viscosity of H₂O+NaCl+CO₂

We have found three literature sources where effects of CO₂ dissolution on brine viscosity were investigated (Bando et al., 2004; Fleury and Deschamps, 2008; Kumagai and Yokoyama, 1999). The study by Kumagai and Yokoyama (1999) is not relevant here because their temperature range is too low (<5° C). The authors Bando et al. (2004) measured the viscosity of brine solutions with dissolved CO₂ at *P-T* ranges of 100-200 bar and 30°-60° C, respectively, at a mass fraction of NaCl between 0 to 0.03. Fleury and Deschamps (2008) studied the effect of dissolved CO₂ on the viscosity of three NaCl solutions covering the range of salinity usually encountered in potential CO₂ storage geological formations. They showed experimental data for viscosity variations in brine solutions for CO₂ dissolution at 35° C and 85 bar, and found viscosity proportional to the mole fraction of CO₂. They made a comparative study of the temperature dependence up to 100° C, with and without dissolved CO₂, with respect to their measured data at 35° C. They presented a model for CO₂ dissolution, but reported that the deviation of calculated results can be even on the order of 10%. Therefore, to reduce error and combine with the data by Bando et al., (2004) we attempted to find a new correlation. Our new modified model is,

$$\mu_{H_2O+NaCl+CO_2} = \mu_{H_2O+NaCl} \left(1 + 4.65x_{CO_2}^{1.0134} \right) \quad (5)$$

$\mu_{H_2O+NaCl}$ can be measured from previous discussion at the same temperature and pressure of $\mu_{H_2O+NaCl+CO_2}$. Comparisons of calculated data from this formulation to the experimental data are shown in Figure 4 and 5 for different temperatures and molalities. The divergences are very scattered, and the error is relatively high (~7%) for some data points. This is because calculated data from the formulations of Fleury and Deschamps (2008), and Bando et al. (2004) differ by 11% at the same conditions, and the uncertainty of their experimental values is around 2-3%. Still, we can recommend that Eq. 3 can be used with confidence for the mentioned *P-T* range based upon the authors' findings that pressure dependence on the viscosity of brine solutions can be ignored and the effect of dissolved CO₂ is not temperature dependent.

Figure 4. Deviation between literature and calculated data by Eq. 5.

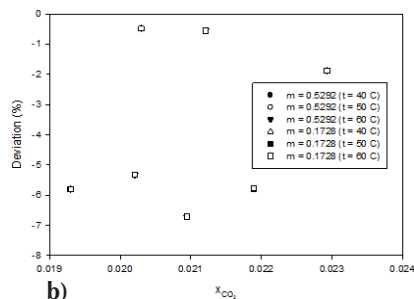


Figure 5. Deviation between literature and calculated data by Eq. 5.

6. Viscosity of Sea Water

In addition to brine water, we also want to propose viscosity model for saline water of typical sea water compositions (Millero et al., 2008). Practically, the salinity (S) of sea water is considered to be 35 (35.16504) gkg^{-1} . A brief list of sea water components is shown in Table 4 adapted from Leyendekkers (1979a).

Table 4. Sea water components ($S = 35$).

Salt, i	$m_{i,s}$ (mol/kg water)	$x_i = \frac{m_i}{\sum m_i}$
NaCl	0.424310	0.79644
Na ₂ SO ₄	0.029245	0.054895
NaHCO ₃	0.002418	0.004539
KCl	0.009412	0.017666
KBr	0.000854	0.001602
MgCl ₂	0.055211	0.103630
CaCl ₂	0.010707	0.020098
SrCl ₂	0.000093	0.000173
H ₃ BO ₃	0.000436	0.000139

A comprehensive literature review was carried out on the viscosity modeling of sea water (Chen et al., 1973; Horne and Johnson, 1966; Isdale et al., 1972; Kobayashi and Nagashima, 1985; Leyendekkers, 1979a, b; Matthaus, 1972; Phang, 1976; Sharqawy et al., 2010; Stanley and Batten, 1969). Most of the studies concentrated on viscosity measurements at atmospheric pressure for different salinity and temperature ranges. Stanley and Batten (1969) performed measurements of viscosity of 3.5% salinity IAP0 (International Association of Physical Oceanography) standard sea water for the pressure range up to 1400 bar and temperatures from 0° to 30° C. Kobayashi and Nagashima (1985) presented viscosity data of synthetic standard sea water for the temperature range 0°-100° C and for pressures up to 400 bar. Their study reveals that above 20° C the effect of pressure can be ignored completely. The pressure dependence on the viscosity of sea water is similar to that of pure water; temperature dependence is not, however. Based upon the experimentally measured data

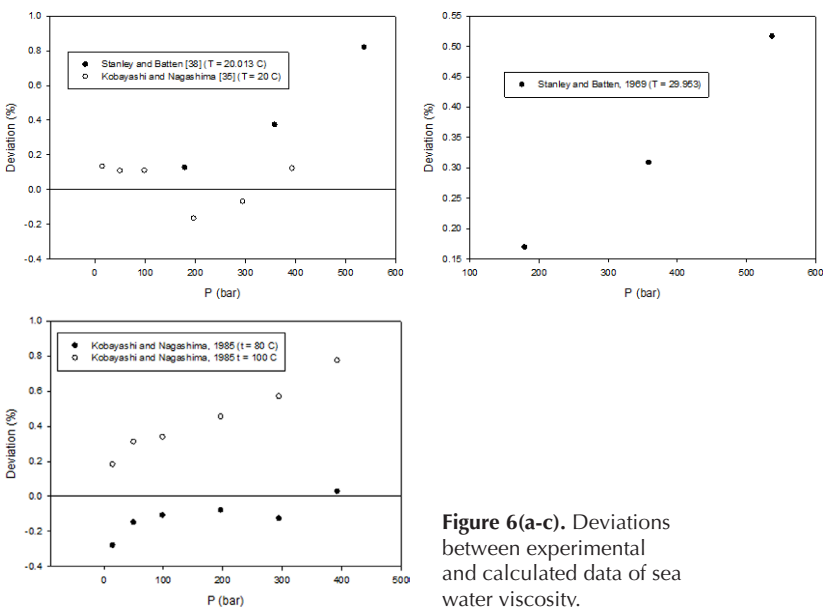


Figure 6(a-c). Deviations between experimental and calculated data of sea water viscosity.

and the viscosity values computed from the models proposed in the literature, our formulation is exactly the same as Eq. 1, but with different coefficients. The new values are shown in Table 5. The errors of computed results by our model are shown in Figure 6(a-c). The results are satisfactory, as deviations lie within 0.9%.

Table 5. Coefficients of Eq. 1 for sea water viscosity.

i	a	b	c	d
0	2.27027348e+01			-2.08076250e-01
1		1.09512918e+04	9.89379527e-03	2.02116962e-02
2		9.59424756e+08	4.38767404e-02	-2.80171705e-04
3		-8.60130112e+08	4.41842469e-02	1.19302430e-06

7. Effects of Dissolved CO₂

In this section we will discuss effect of dissolved CO₂ on the viscosity of the aqueous phase based upon the established models. Figure 7(a-c) show computed viscosity values of H₂O (pure water), H₂O+NaCl (brine water), H₂O+CO₂, H₂O+NaCl+CO₂, and sea water with respect to temperatures at three different pressures (50, 200, and 600 bar), at molality $m = 1$, and CO₂ mass fraction $x_{\text{CO}_2} = 0.02$. The mass fraction was chosen within the solubility

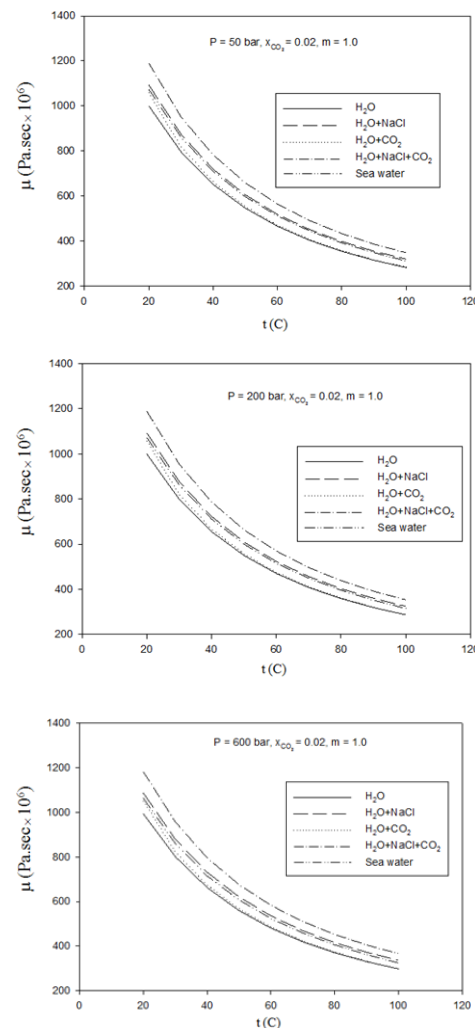


Figure 7(a-c). Viscosity of aqueous solution.

range of CO₂ in pure and brine water (Islam and Carlson, 2012b; Spycher and Pruess, 2005). From these figures it is observed that at lower temperatures (<40° C) the effect of CO₂ dissolution on the viscosity of pure water is relatively high, and tends to diminish as temperature increases. For instance, at $t = 20^\circ \text{C}$, viscosity of water with 2% (by weight) dissolved CO₂ is around 6% higher than pure water, while at 100° C viscosity is 0.6% higher. This finding is consistent with the results by Kumagai and Yokoyama (1999). The phase equilibria and density behavior of aqueous solutions with dissolved CO₂ is similar (Spycher et al., 2003).

Neglecting CO₂'s presence in brine water will warrant large error because, as seen in the figures, at any particular temperature and pressure the viscosity of brine water having 1 molal NaCl with 2% (by weight) dissolved of CO₂ is more than 8% higher than with no CO₂. This percentage will increase with the concentration of NaCl. The phase equilibrium of CO₂ in brine is also consistent with this phenomena (Spycher and Pruess, 2005). The effect of CO₂ in sea water is not tangible because no such study in the literature was found and therefore, no model could be developed.

8. Viscosity of Gaseous Phase

It is well known that, in the gaseous phase, we can assume water composition as zero (Spycher et al., 2003) within the temperature range of geologic sequestration. Therefore, viscosity computations of the effects of water in the gaseous phase can simply be calculated as the viscosity of pure CO₂. Hassanzadeh et al. (2008) have showed this calculation very well.

Concluding Remarks

We have observed one common feature for the models proposed that percentage of deviation remains same at any particular temperature and pressure, irrespective of NaCl molality or dissolved CO₂ composition. This provides confidence in using our formulations. Literature values were reproduced with less than 1% deviation, except in some cases where high experimental uncertainty was reported and the computed values from the earlier models differed around 10%. All possible combinations of viscosity models for the simulation of geologic sequestration of CO₂ are discussed, and the effects of dissolved CO₂ are narrated elaborately. Because the models have fewer parameters, the computational efficiency is also improved.

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Nomenclature

t	Temperature (°C)
T	Temperature (K)
P	Pressure (bar, if not specified)
μ	Dynamic viscosity (micro Pascal.sec)
ρ	Density (kg/m ³)

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