# Coupling Miscible Flow and Geochemistry for Carbon Dioxide Flooding into North Sea Chalk Reservoir

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Abstract: As an effective method to cope with green-house gas emission, and to enhance oil recovery, injection of carbon dioxide (CO<sub>2</sub>) into oil reservoirs has obtained increasing attentions. The flooding process involves complex phase behavior among oil, brine and CO<sub>2</sub>, and geochemical reaction between CO<sub>2</sub> and rock. In this study, COMSOL Multiphysics was first applied to simulating two flooding processes with known analytical solutions to validate the implementation method. Then, a model to simulate miscible  $CO_2$ injection with geochemical reaction in the aqueous phase was established with COMSOL. After validating the model using a finite difference solver for threephase CO<sub>2</sub> flooding, the model was applied to simulating CO<sub>2</sub> flooding process in North Sea chalk both on reservoir scale and laboratory scale.

**Keywords:** miscible multiphase flow, EOR, geochemical reaction.

# 1. Introduction

Geological storage of  $CO_2$  in either aquifer or oil reservoirs is considered to be one of the most efficient ways to reduce  $CO_2$  emission and mitigate global warming. Among the two, storage of  $CO_2$  in oil reservoirs is easier to implement thanks to the plentiful experience accumulated in the oil industry. The process also provides additional benefit by enhancing the oil recovery. The world's largest  $CO_2$  EOR is carried out in Permian basin, USA, with the first project started in 1972. There are more than 23 mt $CO_2/yr$  delivered to more than 50 active projects in the basin, and at the same time, more than 145,000 bpd of incremental oil produced<sup>1</sup>.

The CO<sub>2</sub> flooding process involves complex interaction between oil, water and CO<sub>2</sub> in the coexisiting three fluid phases, such as mutual solubility, change of acidity, and oil swelling, and geochemical reactions between aqueous phase and rock. The three-phase fluid flow and

geochemical process also influence each other. The former determines how solutes are transported; the latter changes the flow by changing porosity and permeability due to mineral dissolution and precipitation. Therefore, there is a strong coupling between transport and geochemical reaction.

The reservoir simulators for CO<sub>2</sub> flooding and the geothermal simulators for CO<sub>2</sub> sequestration usually have different focuses. The former focuses on three-phase flow and phase equilibrium<sup>2</sup>, while the latter focuses on geochemical reaction in the aqueous phase<sup>3</sup>, with only two phases (gas and aqueous) flow included. In this study, it is investigated how to simulate CO<sub>2</sub> flooding including both threephase flow and geochemical reaction by using Multiphysics (hereafter called COMSOL COMSOL). COMSOL has been used to simulate EOR process before but only for immiscible flow<sup>4</sup>. Therefore, to test if COMSOL can be applied to situations with component exchange between phases, two simple EOR processes, gas flooding and polymer flooding, were tested first. Then, we established a model on miscible  $CO_2$ flooding with consideration of mineral dissolution and change of geophysical properties. Finally, the model was applied to simulating  $CO_2$ flooding in chalk both on reservoir scale and lab scale.

# 2. Numerical tests with COMSOL Multiphysics

In this section, two numerical tests are used to evaluate the performance of COMSOL in modeling miscible flow and transport-reactive flow. The two examples are taken from the existing publications<sup>5,6</sup>. In all the following description, we use subscripts i and j to represent components and phases, respectively. The two examples are input into the COMSOL, by using the PDE Modes and the coefficient form.

# 2.1. Gas flooding

In gas flooding, component separation happens while flowing. The transfer of components between flowing phases strongly influences displacement performance. A 1-D two-component gas/oil displacement is studied here. The transport equation for the gas component and the saturation equation are,

$$\phi \frac{\partial}{\partial t} (S_g \rho_g x_{g,g} + S_o \rho_o x_{g,o}) + \\ \frac{\partial}{\partial r} (x_{g,g} \rho_g u_g + x_{g,o} \rho_o u_o) = 0$$
(1)

$$S_g + S_o = 1 \tag{2}$$

where,  $x_{g,j}$  is the molar fraction of gas component in the gas (g) or oil (o) phase,  $S_j$  is the saturation,  $\rho_j$  is the molar density.  $\phi$  is the porosity. Without consideration of capillary pressure, phase flow velocity,  $u_j$ , becomes,

$$u_j = -\frac{kk_{rj}}{\mu_j} \frac{\partial P}{\partial x} \tag{3}$$

in eq. (3), k is the absolute permeability,  $k_{r,i}$  and  $\mu_j$  are the relative permeability and viscosity. P is the pressure. The flow velocity can be written in terms of fractional flow functions,  $f_j$ , defined by,

$$u_j = f_j u = f_j (u_o + u_g) \tag{4}$$

in eq.(4), u is the total velocity. The relative permeability for oil and gas can be expressed as, for gas,

$$k_{rg} = 0, (S_g < S_{gc})$$

$$k_{rg} = \left(\frac{S_g - S_{gc}}{1 - S_{gc} - S_{or}}\right)^2, (S_{gc} < S_g < 1 - S_{or}) (5)$$

$$k_{rg} = 1, (S_g > 1 - S_{wr})$$

for oil,

$$k_{ro} = 0, (1 - S_g < S_{or})$$

$$k_{ro} = \left(\frac{1 - S_g - S_{or}}{1 - S_{gc} - S_{or}}\right)^2, (S_{gc} < S_g < 1 - S_{or})$$

$$k_{ro} = 1, (1 - S_g > 1 - S_{gc})$$
(6)

by substituting eqs. (3), (5) and (6) into eq.(4), the fractional flow for gas phase  $f_{g}$  becomes,

$$f_g = 0, (S_g < S_{gc}) f_g = \left(\frac{(S_g - S_{gc})^2}{(S_g - S_{gc})^2 + (1 - S_g - S_{or})^2/M}\right) (7) (S_{gc} < S_g < 1 - S_{or}) f_g = 1, (S_g > 1 - S_{or})$$

in eq. (7),  $S_{gc}$  is the critical gas saturation,  $S_{or}$  is the residual oil saturation. M is the viscosity ratio between oil and gas. For this example, we assume  $S_{gc} = 0.05$ ,  $S_{or} = 0.1$  and M = 2. Under the assumption that the molar density of gas component,  $\rho_{cg}$ , is constant in two phases and we have<sup>5</sup>,

$$\rho_{cg}c_{g,j} = \rho_j x_{g,j} \tag{8}$$

where  $c_{g,j}$  is the volume fraction of gas component in different phases. By substituting eq. (4) and (8) into equation (1), we obtain

$$\phi \frac{\partial}{\partial t} (S_g c_{g,g} + S_o c_{g,o}) + \\ v \frac{\partial}{\partial c} (c_{g,g} f_g + c_{g,o} f_o) = 0$$
(9)

by introducing dimensionless parameters,

$$\tau = \frac{u_{inj}t}{\phi L}, \, \zeta = \frac{x}{L}, \, u_D = \frac{u}{u_{inj}} \tag{10}$$

and with  $u_D = 1$ , eq. (9) becomes,

$$\frac{\partial N_g}{\partial \tau} + \frac{\partial F_g}{\partial \zeta} = 0 \tag{11}$$

where  $N_g$  is the overall volume fraction of gas component,  $N_g = S_g c_{g,g} + S_o c_{g,c}$ , and  $F_g$  is the overall fractional volumetric flow of gas component,  $F_g = f_g c_{g,g} + f_o c_{g,c}$ . To compare the results with the analytical solution, P is assumed to be constant, and correspondingly  $c_{g,g}$ and  $c_{g,c}$  are constant in the isothermal process. We assume  $c_{g,g} = 0.95$  and  $c_{g,o} = 0.2$ 

An artificial diffusion coefficient D = 1E - 4 is introduced to avoid oscillation. The initial condition for  $N_g$  is 0.05, the left boundary condition is  $N_g = 0.975$ , and the right boundary condition is Neumann type condition with q = g = 0 (q and g are parameters in COMSOL).



Figure 1 Comparison of analytical solution and COMSOL solution in gas flooding

As shown in fig.1, the results from COMSOL agree well with those from analytical solution at  $\tau = 0.6$ .

#### 2.2. Polymer flooding

In polymer flooding, polymer is added to water to increase its viscosity so as to mobilize more oil. However, the polymer can be adsorbed on rock surface, and the decreasing concentration of polymer in water will affect the flow of water phase. The process is characterized by the coupling between adsorption and flow transport.

The transport equations for 1-D polymer flooding are

$$\phi \frac{\partial S_w}{\partial t} + u \frac{\partial f_w}{\partial x} = 0$$

$$\phi \frac{\partial}{\partial t} (S_w c + c_s) + u \frac{\partial}{\partial x} (f_w c) = 0$$
(12)

where c is the concentration of polymer in water phase.  $c_s$  is the concentration of adsorbed polymer. By using eq. (10) with  $u_D = 1$ , eq. (12) becomes

$$\frac{\partial S_w}{\partial \tau} + \frac{\partial f_w}{\partial \zeta} = 0$$

$$\frac{\partial}{\partial \tau} (S_w c + c_s) + \frac{\partial}{\partial \zeta} (f_w c) = 0$$
(13)

The relative permeabilities of oil and water,  $k_{re}$  and  $k_{ru}$ , are

$$k_{ro} = (1 - \psi)^2 (1 + 2\psi)$$
  

$$k_{rw} = 0.25\psi^2$$
(14)

where  $\psi$  is

$$\psi = \frac{S_w - S_{wr}}{1 - S_{or} - S_{wr}}$$

 $S_{wr}$  and  $S_{or}$  are the residual saturation of water and oil respectively. Here,  $S_{wr}$  and  $S_{or}$  are set to 0.2 and 0.3 respectively. The viscosity of water phase is assumed to be a linear function of c,

$$\mu_w = \mu_w^0 (1 + \beta c) \tag{15}$$

where  $\mu_{u}^{0}$  is the initial water viscosity,  $\beta$  is constant. The viscosity of oil,  $\mu_{a}$ , is constant.  $f_{u}$  can thus be expressed by,

$$f_w = \frac{k_{rw}}{k_{rw} + k_{ro}\mu_w/\mu_o}$$
(16)  
=  $\frac{\psi^3}{\psi^3 + \alpha(1 + \beta c)(1 - \psi)^2(1 + 2\psi)}$ 

where  $\alpha = 4\mu_w^0/\mu_c$ . The equilibrium relation for the polymer adsorption on rock surfaces is often given by the Langmuir isotherm

$$c_s = \frac{NKc}{1+Kc} \tag{17}$$

where N and K are constant.  $\mu_c$  is set to 40cp,  $\mu_u^0$  1cp,  $\beta$  4 L/g, N 0.6 g/L and K 10 L/g.

An artificial diffusion coefficient D = 2E - 3 is also introduced here. The initial and boundary conditions are

$$\tau = 0$$
:  $S_w = 0.6$ ,  $c = 0$  g/liter  
 $\zeta = 0$ :  $S_w = 1.0$ ,  $c = 1.0$  g/liter

Fig. 2 and fig. 3 show that the numerical results from COMSOL agree well with those from the analytical solution at  $\tau = 0.4$ , in terms of both the water saturation,  $S_{u}$ , and the polymer concentration in water phase, c.



Figure 2 Comparison of analytical solution and COMSOL solution in polymer flooding,  $S_{u}$ .



**Figure3** Comparison of analytical solution and COMSOL solution in polymer flooding, c

# **3.** Modeling of CO<sub>2</sub> flooding in North Sea chalk reservoir

# 3.1. Description of the system and equations

When  $CO_2$  is injected into oil reservoirs, three phases are present: oil,  $CO_2$  and water. The water phase contains ions and dissolved  $CO_2$ . The main mineral reaction considered here is

$$CO_2(aq) + H_2O + CaCO_3(mineral) \iff Ca^{2+} + 2HCO_3^-$$

The following assumptions are used:

- 1. This process is isothermal;
- 2. Only CO<sub>2</sub> is dissolved in water;
- 3. Chemical reaction has no influence on the molar density of water phase;
- 4. No capillary pressure is considered;
- 5. Molar density of single component in the different phases is same.

The mass conservation equations are Oil component:

$$\frac{\partial}{\partial t}(\phi S_o \rho_o) + \frac{\partial}{\partial x}(u_o \rho_o) = 0$$
(18)

CO<sub>2</sub> component:

$$\frac{\partial}{\partial t} (\phi S_g \rho_g + \phi S_w \rho_w x_{CO_2,w}) +$$

$$\frac{\partial}{\partial x} (u_w \rho_w x_{CO_2,w} + u_g \rho_g) = -R_m$$
(19)

Water component:

$$\frac{\partial}{\partial t} (\phi S_w \rho_w x_{H_2O,w}) + \\ \frac{\partial}{\partial t} (u_w \rho_w x_{H_2O,w}) = -R_m$$
(20)

Calcium component;

$$\frac{\partial}{\partial t} (\phi S_w \rho_w x_{Ca^{2+},w}) +$$

$$\frac{\partial}{\partial r} (u_w \rho_w x_{Ca^{2+},w}) = R_m$$
(21)

In the above equations,  $\phi$  is the porosity,  $S_j$ (j = o, g, w) are the saturation of oil, gas and brine phase,  $\rho_j$  is the molar density of different phases,  $x_{i,j}$  is the molar fraction of component *i* in the phase *j* (*i* = *CO*<sub>2</sub>, *H*<sub>2</sub>*O*, *Ca*<sup>2+</sup>, *HCO*<sub>3</sub><sup>-</sup>),  $u_j$  is the Darcy velocity as described by eq. (3). The relative permeability  $k_{r_1}$  is

$$k_{ri} = S_i^2 \tag{22}$$

The neutrality condition requires

$$2x_{Ca^{2+},w} = x_{HCO_{3}^{-},u} \tag{23}$$

$$x_{CO_2,w} + x_{Ca^{2+},w} + x_{HCO_3^-,w} + x_{H_2O,w} = 1$$
(24)

The saturations are subject to the constraint  

$$S_w + S_g + S_o = 1$$
 (25)

The porosity change is given by

$$\frac{\partial (1-\phi)\rho_m}{\partial t} = -R_m \tag{26}$$

where  $\rho_m$  is the molar density of mineral.  $R_m$  is the reaction term, and can be expressed by  $R_m = S_b r_m$ , and  $r_m$  is the kinetic reaction term  $a_{CO_2,b}^{-1} a_{Ca^{2+},b} a_{HCO_3,b}^2$ , (27)

$$r_m = k_m A_m \left(1 - \frac{a_{CO_2, b} a_{Ca^{2+}, b} a_{HCO_3^-, b}}{K_{sp}}\right)$$
(27)

where  $k_m$  is the rate constant,  $A_m$  is the specific surface of mineral,  $K_{s_F}$  is the solubility product, and  $a_i$  is the activity for aqueous components. Here we assume the activity of water is 1.

In our system,  $x_{CO_2,u}$ , can be treated as a function of P. Therefore, there are total 8 independent unknowns,  $S_c$ ,  $S_g$ ,  $S_u$ , P,  $x_{H_2O,u}$ ,  $x_{Ca^{2+},u}$ ,  $x_{HCO_3^-,u}$ ,  $\phi$  for the 8 independent equations, eq. (18)-(21), (23)-(26).

#### 3.2. Solution method

There are two approaches to solving the coupled equations: the sequential solution method and the simultaneously solution method, which implies that all equations are solved simultaneously. In the sequential approach, flow equations and chemical-reaction equations are solved separately and sequentially<sup>3</sup>. If iterations are made between two steps until convergence, the approach is called sequential iterative approach (SIA). Otherwise, it is called sequential non iterative approach (SNIA). SNIA is used here to solve the coupled equations. The approach is similar to the one used in the multiphase reactive geochemical simulator, TOUGHREACT<sup>7</sup>.

The solution method is shown in fig. 4. At each time step, pressure equation and flow equations are solved initially. Then, solute transport equation is solved with known pressure and saturation. Finally, with updated chemical reaction term, the porosity is calculated.



Figure 4 SNIA solution method used in the model

#### **Pressure equation and flow equations**

Since the change of  $x_{Ca^{2+},u}$  has minor influence on the calculation of phase saturation  $S_j$ , eqs. (18)-(20) are used as flow equations without consideration of chemical reaction term. By applying eq. (8) and fractional flow functions,  $f_j$ , in a way similar to that in gas flooding, eqs. (18)-(20) become

$$\frac{\partial}{\partial t}(\phi G_o) + u \frac{\partial}{\partial x}(H_o) = 0$$
(28)

$$\frac{\partial}{\partial t}(\phi G_g) + u \frac{\partial}{\partial x}(H_g) = 0$$
 (29)

$$\frac{\partial}{\partial t}(\phi G_w) + u \frac{\partial}{\partial x}(H_w) = 0$$
 (30)

where

$$\begin{split} G_o &= \rho_{co} N_o = \rho_{co} S_c, \\ H_o &= \rho_{co} F_o = \rho_{co} f_c, \\ Gg &= \rho_{cg} N_g = \rho_{cg} (S_g + S_w c_{CO_2,w}), \\ H_g &= \rho_{cg} F_g = \rho_{cg} (f_g + f_w c_{CO_2,w}), \\ G_w &= \rho_{cw} N_w = \rho_{cw} S_w (1 - c_{CO_2,w}), \\ H_w &= \rho_w F_w = \rho_{cw} f_w (1 - c_{CO_2,w}), \end{split}$$

 $\rho_{cc}$ ,  $\rho_{cg}$ ,  $\rho_{cu}$  are the molar density of oil component, CO<sub>2</sub> component, and H<sub>2</sub>O component, respectively. They are constant in the model.  $c_{CO_2,u}$  is the volumetric fraction of CO<sub>2</sub> dissolved in the water. By deleting the molar density in eqs. (28)-(30), and adding them together, we obtain the pressure equation

$$\frac{\partial}{\partial x} \left( \left( \frac{kk_{ro}}{\mu_o} + \frac{kk_{rg}}{\mu_g} + \frac{kk_{rw}}{\mu_w} \right) \frac{\partial P}{\partial x} \right) = 0 \quad (31)$$

Only two flow equations in eqs. (28)-(30) are independent and eqs. (29)-(30) are chosen to be the flow equations

#### Solute transport equation

In the system described here, since  $c_{CO_2,u}$  is constant at a certain pressure, molar fraction of CO<sub>2</sub> in water  $x_{CO_2,u}$  and the updated molar density of water phase are,

$$\rho_w = \rho_{cg} c_{CO_2,w} + \rho_{cw} (1 - c_{CO_2,w})$$
(32)

 $\rho_{cg}c_{CO_2,w} = \rho_w x_{CO_2,u}$ (33) by substituting eqs. (27) and (23) into eq.(21), we obtain

$$\frac{\partial}{\partial t}(\phi S_w \rho_w x_{Ca^{2+},w}) + \frac{\partial}{\partial x}(u_w \rho_w x_{Ca^{2+},w}) =$$

$$S_w k_m A_m (1 - \frac{2\rho_w^2 x_{Ca^{2+},b}^3}{x_{COa,b} K_{sa}})$$
(34)

where the activity coefficient of components are assumed to be 1 and thus the kinetic reaction term can be expressed by molar concentration of components.

#### **Porosity equation**

With the updated reaction term,  $R_m$ , porosity can be solved by eq. (26).

Following the solution method in fig.4, the above equations are implemented into COMSOL by using the PDE Modes, the coefficient form and solved by the segregated solver.

#### 3.3. Validation of the model

A finite difference code for 3-phase immiscible or miscible flooding without chemical reaction is used to validate the model. By using eq.(10), eqs. (29)-(30) become dimensionless:

$$\frac{\partial N_g}{\partial \tau} + \frac{\partial F_g}{\partial \zeta} = 0 \tag{35}$$

$$\frac{\partial N_w}{\partial \tau} + \frac{\partial F_w}{\partial \zeta} = 0 \tag{36}$$

The initial condition is Ng = 0,  $N_w = 0.4$ , and the boundary condition is  $N_g = 1$ ,  $N_w = 0$ . The parameters are  $c_{CO_2,w} = 0.05$ ,  $\mu_g = 0.1cp$ ,  $\mu_o = 0.8cp$ , and  $\mu_w = 1cp$ .



Figure 5 Three-phase flooding without reaction

If the injection velocity is constant, the results from COMSOL agree well with those from the 1-D solver, as shown in fig.5 at  $\tau = 0.2$ . As before, a diffusion coefficient D = 1E - 3 has been introduced to avoid oscillation.

### 3.4. Simulation of reservoir scale flooding

The model is used to simulate field scale  $CO_2$  flooding in a 1-D chalk reservoir. The pressure and temperature are 100 bar and 50 °C respectively. The oil in the reservoir is assumed to be n-decane. The simulation parameters are listed in table 1. The initial and boundary conditions are listed in table 2.

Table 1 Values of constants									
Property	Unit	Value	Property	Unit	Value				
$c_{CO_2,u}$		0.02	$k_m$	$\frac{mol}{(m^2s)}$	1.6E-9				
$\mu_c$	cp	0.7	$A_m$	$\frac{m^2}{m^3}$	10000				
$\mu_u$	cp	0.6	$k_{sp}$		1E-5				
$\mu_g$	cp	0.06	k	$m^2$	3E-15				
$\rho_{cc}$	$\frac{mol}{m^3}$	4900	$\phi$		0.3				
$\rho_{cu}$	$\frac{mol}{m^3}$	54000	$\rho_m$	$\frac{mol}{m^3}$	27100				
$\rho_{cg}$	$\frac{mol}{m^3}$	16000							

Table 1 Values of constants

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	Initial	Boundary(L)	( <b>R</b> )
P	100bar	120bar	100bar
$G_{g}$	$3.792 \frac{mol}{m^3}$	$16000 \frac{mol}{m^3}$	Same as
$G_{u}$	$6467.25 \frac{mol}{m^3}$	$0 \frac{mol}{m^3}$	injection
$x_{ca^{2+},w}$	0.0001	0	-
$\phi$	0.3	0	

The model has been used to simulate the  $CO_2$  flooding in reservoir for 400 days. Fig. 6 shows how the gas saturation profile moves with time. The gas front breaks through after 500 days.



Figure 6  $S_4$  from 100 to 400 days

Fig. 7 shows the profile of molar concentration of calcium,  $x_{Ca^{2+},u}$ . It reaches equilibrium even at early stage, indicating that the dissolution of calcite for reservoir scale flooding can be treated as equilibrium reactions.



Figure 7  $x_{Ca^{2+},u}$  from 100 to 400 days

#### 3.5. Simulation of lab scale flooding

As comparison, the model was also used to simulate CO<sub>2</sub> flooding in a chalk core in the lab experiment. The core length is 120 mm, and the diameter is 1.5 inch (38.1mm). The injection rate under experimental condition is 1 cc/min, corresponding to an injection velocity of 1.5E-6 m/s. Fig. 8 shows the influence of kinetic reaction on the increase of  $x_{Ca^{2+},u}$ .  $x_{Ca^{2+},u}$  increases with time but its value is far below the equilibrium value, indicating that the dissolution of calcite is limited for short injection time at the Lab experiment.



Figure 8  $x_{Ca^{2+},u}$  from 0.5 to 2 hours along the core

Fig. 9 illustrates the influence of reactive surface area,  $A_m$ . For  $A_m=1E+6$  m<sup>2</sup>/m<sup>3</sup>,  $x_{Ca^{2+},u}$  almost reaches equilibrium, while for  $A_m=1E+4$  m<sup>2</sup>/m<sup>3</sup>, the change of  $x_{Ca^{2+},u}$  is almost negligible.



Figure 9 Effect of  $A_m$  on  $x_{Ca^{2+},u}$  after 1 hour



**Figure 10** Effect of  $A_m$  on  $\phi$  after 1 hour

Fig.10 shows the porosity change at different  $A_m$ . The change is almost negligible even at near equilibrium condition.

# 4. Conclusions

Using SNIA, we have developed a model in COMSOL for miscible  $CO_2$  flooding with dissolution reaction in the aqueous phase. It seems that COMSOL is able to provide reasonable results for this type of reactive transport problem by using the segregated solver. The model will be useful to study  $CO_2$  flooding in chalk reservoir and  $CO_2$  sequestration process. The phase equilibrium between  $CO_2$  and oil can be added although it is not included by far. The geochemical model should be extended to include more reactions in order to simulate real  $CO_2$  sequestration scenarios. Other solution method can also be tried in the future.

#### 5. Acknowledgement

The authors are grateful to Lars Gregersen, at COMSOL A/S, Denmark, for his help and suggestions. This study is carried out under the project "Enhanced Oil Recovery through  $CO_2$  Utilization" funded by the Danish National Advanced Technology Foundation.

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