

## Property impacts on Carbon Capture and Storage (CCS) processes

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### Abstract:

The knowledge of thermodynamic and transport properties of CO<sub>2</sub>-mixtures is important for designing and operating different processes in carbon capture and storage systems. A literature survey was conducted to review the impact of uncertainty in thermos-physical properties on the design and operation of components and processes involved in CO<sub>2</sub> capture, conditioning, transport and storage. According to the existing studies on property impacts, liquid phase viscosity and diffusivity as well as gas phase diffusivity significantly impact the process simulation and absorber design for chemical absorption. Moreover, the phase equilibrium is important for regenerating energy estimation. For CO<sub>2</sub> compression and pumping processes, thermos-physical properties have more obvious impacts on pumps than on compressors. Heat capacity, density, enthalpy and entropy are the most important properties in the pumping process, whereas the compression process is more sensitive to heat capacity and compressibility. In the condensation and liquefaction process, the impacts of density, enthalpy and entropy are low on heat exchangers. For the transport process, existing studies mainly focused on property impacts on the performance of pipeline steady flow processes. Among the properties, density and heat capacity are most important. In the storage process, density and viscosity have received the most attention in property impact studies and were regarded as the most important properties in terms of storage capacity and enhanced oil recovery rate. However, for physical absorption, physical adsorption and membrane separation, there has been a knowledge gap about the property impact. In addition, due to

the lack of experimental data and process complexity, little information is available about the influence of liquid phase properties on the design of the absorber and desorber for chemical absorption process. In the CO<sub>2</sub> conditioning process, knowledge of the impacts of properties beyond density and enthalpy is insufficient. In the transport process, greater attention should focus on property impacts on transient transport processes and ship transport systems. In the storage process, additional research is required on the dispersion process in enhanced oil recovery and the dissolution process in ocean and saline aquifer storage.

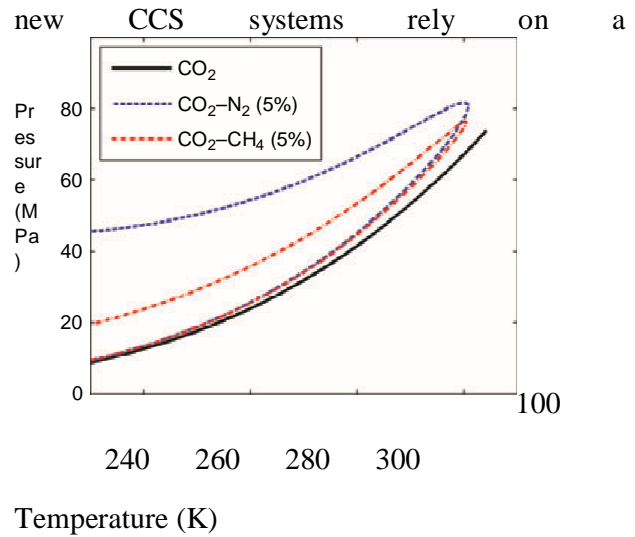
### 1. Introduction

Technology for CO<sub>2</sub> capture and storage (CCS) is expected to play an important role to mitigate global warming. This is, for instance, shown in the Blue map presented by IEA (International Energy Agency) in their report on energy technology perspectives, where CCS accounts for one-fifth of the total reduction of CO<sub>2</sub> emissions (IEA, 2011). A prerequisite for development, design, and improvement of CCS technologies is the ability to accurately predict the behavior of the fluids involved. It is well known that CO<sub>2</sub>-rich mixtures behave very differently from natural gas, flue gas, and other compositions, which properties are well documented in the literature. In addition, it has been shown that small amount of impurities can substantially change the properties of the mixture, compared to pure CO<sub>2</sub>. This is illustrated in Figure 1, where the phase envelopes of CO<sub>2</sub>CH<sub>4</sub> and CO<sub>2</sub>N<sub>2</sub> mixtures are compared to the vapor-liquid line of pure CO<sub>2</sub>. Different impurities give the CO<sub>2</sub> mixture different properties. As shown in Figure 1, a CO<sub>2</sub>

mixture with 5% methane has a narrower phase envelope than that with nitrogen. Knowledge of the behavior of CO<sub>2</sub> mixtures will allow design of safe, economical and efficient processes, and safe operation of the CCS technology.

### 1.1 Importance of properties to CCS

The presence of impurities such as O<sub>2</sub>, H<sub>2</sub>S, CH<sub>4</sub>, and N<sub>2</sub> affects the overall thermophysical properties of CO<sub>2</sub> mixtures substantially. In general, the critical points shift to higher pressures with impurities in CO<sub>2</sub>; moreover, the phase change does not happen at a constant temperature or pressure, but ranges in temperature and pressure, as shown in Figure 1. This increases the probability to encounter two phases in the CCS process. The density represents another important property for the design and development of CCS technologies. For instance, a small amount of CH<sub>4</sub> reduces the CO<sub>2</sub> density significantly. Therefore, for the same pressure, a higher compressibility is expected for CO<sub>2</sub>/CH<sub>4</sub>. This will affect the pipeline flow, e.g. the volume requirement to maintain the reservoir pressure [1-3]. The transport properties of CO<sub>2</sub>-rich mixtures are important properties closely connected to heat, momentum, and mass transfer in CCS. Accurately predicting the pressure drop in pipelines requires viscosity, which typically has not yet been measured for CO<sub>2</sub> mixtures in the liquid phase area. The CCS community is in agreement about the importance of transport properties in the development, design, and simulation of amine-based chemical absorption columns. It is essential to provide accurate predictions of viscosities, thermal conductivities, and diffusion coefficients (Hoff et al., 2004). The surface tension of CO<sub>2</sub> mixtures determines the flow pattern of two-phase fluids in process equipment. It is fair to claim that the development and technical breakthrough of



**Figure 1.** Impact of impurities on the phase envelopes of CO<sub>2</sub> mixtures predicted with the Peng–Robinson equation of state with regressed interaction parameters. Better understanding of the thermophysical properties of CO<sub>2</sub> mixtures.

### 1.2 Current status about experimental studies of properties

The properties of pure CO<sub>2</sub> have been studied for decades, and there are well-established databases for thermodynamic and transport properties. However, for CO<sub>2</sub> mixtures, experimental activities have mainly focused on compositions and conditions relevant to processing natural gas resources and to enhanced oil recovery (EOR). Hence, there is a substantial amount of data available for mixtures of CO<sub>2</sub>/H<sub>2</sub>O, CO<sub>2</sub>/N<sub>2</sub>, CO<sub>2</sub>/CH<sub>4</sub>, and CO<sub>2</sub>/H<sub>2</sub>S at a wide range of temperatures and pressures [4-6]. There are also some data available for ternary CO<sub>2</sub> mixtures, such as CO<sub>2</sub>/CH<sub>4</sub>/N<sub>2</sub> and CO<sub>2</sub>/N<sub>2</sub>/O<sub>2</sub>. For other impurities important for CCS processes, such as O<sub>2</sub>, Ar, and SO<sub>2</sub>, the experimental data are limited or non-existent.

Except for CO<sub>2</sub>/H<sub>2</sub>O/(NaCl) and CO<sub>2</sub>/amine/H<sub>2</sub>O mixtures, there are no available measurements regarding the transport properties of liquid-phase mixtures. Moreover, there are few data available for the transport properties at high temperatures and pressures. In addition, there are no data

available for the diffusion coefficients of the CO<sub>2</sub>/CH<sub>4</sub> mixture, which may be important in storage calculations. The technology for measurements of sufficient accuracy for transport properties was only available after 1970. Nevertheless many experimental data have been published before this and hence poor accuracy is expected. However, many theoretical models have been fitted to these data (Li *et al.*, 2011a).

### 1.3 Objectives of the article

This article contains an overview about the available experimental data and theoretical models for the main properties of CO<sub>2</sub> mixtures. The purpose of this work is to summarize and evaluate the available experimental data, identify knowledge gaps, and investigate the models that have been proposed and tested in the literature [7-9]. The work will establish the state of the art and provide suggestions for future research on the properties of CO<sub>2</sub> mixtures.

## 2. CO<sub>2</sub> capture by chemical absorption

### 2.1. Process description and related properties of chemical absorption

#### 2.1.1. Process description of chemical absorption

Chemical absorption using amine solvents is widely used and commercially mature for separating CO<sub>2</sub>. It can be applied to gas with a low CO<sub>2</sub> concentration due to its high reaction rate with CO<sub>2</sub>. The chemical absorption process is illustrated in

Fig. 2. An absorber, a heat exchanger, a desorber and a reboiler are the key pieces of equipment. The flue gas is passed into the absorber at the bottom and comes into contact with the solution such as aqueous monoethanolamine (MEA) injected from the top. The separation of CO<sub>2</sub> from the flue gas occurs as mass transfer is enhanced by chemical reaction. The CO<sub>2</sub> rich solvent leaves the absorber at the bottom and enters through the top of the desorber where it flows downward and comes into contact with the stripping steam generated in the reboiler. Most of the CO<sub>2</sub> is

desorbed inside the desorber and partly in the reboiler. The desorbed CO<sub>2</sub> leaves the top of the desorber at a high temperature and most of water vapor is condensed in the condenser [10-12]. The regenerated lean solution is then returned to the absorber through the rich/lean heat exchanger and cooling unit to lower its temperature. Solvent and water make-up are passed to the lean solution to keep the circulating solvent flow rate constant and to maintain its amine concentration.

#### 2.1.2. Key components and related properties of chemical absorption

To achieve a high recovery rate, large volumes of the absorber and desorber are typically required. Consequently, the regeneration process consumes a significant amount of thermal energy. Therefore, the sizing of the absorber/desorber tower and the operating condition and energy requirement of the reboiler are important issues for process application. Designing the columns, such as determining their height and diameter and their packing type and flooding velocities, requires knowing the properties of fluids. According to the review on modeling CO<sub>2</sub> capture by chemical absorption, thermo-physical properties are crucial to achieve accurate simulations of pressure drop, CO<sub>2</sub> recovery rate and purity.

##### 2.1.2.1. Columns diameter.

CO<sub>2</sub> absorption and desorption are generally operated in a counter-current flow. The diameters of the absorber and desorber columns can be estimated using a similar methodology. The column diameter is determined by the properties, flow rate and packing specification. The flooding condition can be determined by the maximum gas velocity because the down-flowing liquid resistance and the column pressure drop increase with increasing gas velocity. The diameter design of the packing column is typically based on 70–80% gas velocity or flux. The flooding velocity can be retrieved from the gas flux at flooding when the mass flow rate of gas phase is known. From the well-known generalized

pressure drop correlation (GDPC) for packed columns by, the gas flux at flooding is calculated from the packing factor, the flooding capacity parameter, the density from both phases, and the liquid phase viscosity. Moreover, the flow parameter is a function of mass flow rate and the density of the fluids [13-15]. The same properties are used to modify the GDPC correlation proposed by Strigle for a more accurate design of the new generation of packing. The pressure drop of the two-phase flow in a packed column can be obtained from GDPC graphs when the flow parameter and the capacity parameter are calculated. The densities of both phases are required to calculate the flow parameter. For capacity parameter calculation, both phase density and liquid phase viscosity are required. Numerical methods are also used to calculate the pressure drop, such as the methods proposed. According to the pressure drop inside the desorber varies with the geometric surface and the void volume of the column packing, the gas load factor, the wall factor and the resistance coefficient. Because the free-cross section for the gas flow is reduced by the column holdup, and the surface structure changes as a result of coating with the liquid film, the column holdup must be considered in performing the pressure drop calculation. To obtain the pressure drop by this method, the density and viscosity of both phases are required. To use the method proposed by, the liquid phase density and viscosity are required to calculate the liquid loading factor, while the gas phase density is required to calculate the gas loading factor. The gas loading factor is used to calculate the pressure drop of the dry packing bed. The pressure drop of the wet packing can be estimated subsequently from the pressure drop of the dry packing bed together with the liquid loading factor.

#### 2.1.2.2. Packing height.

To design the packing height of the absorber and desorber columns, electrolyte thermodynamics, chemical reactions, and heat and mass transfer across the gas-liquid interface and in the bulk gas

and liquid phases must be considered. Mathematical models to design the packing height have been developed in several studies. In general, to calculate the CO<sub>2</sub> absorbed by amine solution, the gas/liquid-phase mass transfer coefficients, the effective interfacial area, the enhancement factor, and the phase equilibrium of the system are required. Onda et al. developed the first and still widely used correlations for mass transfer and effective interfacial area based on wetted wall columns for random packing. The gas phase mass transfer coefficient is directly proportional to the diffusivity and density, but inversely proportional to the viscosity. For liquid phase, the effective interfacial area is considered for completing the mass transfer estimation. This parameter is obtained from a detailed investigation of the effect of hydrodynamics and liquid physical properties, namely density, viscosity and surface tension. Therefore, the liquid phase mass transfer coefficient is directly proportional to the diffusivity, density and surface tension, but inversely to the viscosity. Several correlations proposed by other studies for random packing show similar relationships between mass transfer correlations and the fluid properties. For structured packing, in the correlations proposed by, the gas phase mass transfer coefficients vary with both gas and liquid phase properties. They are directly proportional to the diffusivity and densities of the gas phase, but inversely proportional to the liquid phase density and the viscosity of both phases. Moreover, the liquid phase coefficient is directly proportional to the diffusivity, but inversely proportional to the viscosity and the density.

### 3. CO<sub>2</sub> capture by physical absorption

#### 3.1. Process description and related properties of physical absorption

##### 3.1.1. Process description of physical absorption

Physical absorption is also a leading commercial option for CO<sub>2</sub> capture technology, particularly in natural gas purification and pre-combustion capture. Unlike chemical absorption, the

absorption mechanism of physical absorption occurs in one step, which dissolves CO<sub>2</sub> into the liquid solvent. The main advantage of physical absorption is that the absorption is not limited by the fixed stoichiometry of the reaction as in the case of chemical absorption. The processes are proven to be efficient and economical when operated at higher pressures and lower temperatures. Compared with chemical absorption, less energy is required for physical absorption, and it is also more profitable to treat the feed gas with a high CO<sub>2</sub> concentration. Physical absorption mainly consists of two steps: CO<sub>2</sub> absorption and solvent regeneration by desorption and dehydration. Due to the high vapor pressure of the solvent, the absorption process must be operated at a low temperature and a high pressure to avoid significant solvent loss at a high temperature while maintaining the absorption capacity. The high pressure syngas is first cooled to the design absorption temperature before being absorbed to the solvent that has a high solubility for CO<sub>2</sub> in the absorber column. The reaction of CO<sub>2</sub> absorption in the physical solvent is an exothermic reaction. Therefore, the rising of solvent temperature inside the column must be controlled. The CO<sub>2</sub> loaded solvent can be regenerated with three different methods: flash at reduced pressure, as shown in Fig. 3; stripping; or distillation. The process used depends on the required purity of the treated gas.

### 3.1.2. Key components and related properties of physical absorption

The key components of physical absorption are the flue gas absorber, regeneration units, and the refrigeration system. The process power and energy demand are located primarily in the refrigeration system, the feed gas compressor and the solvent pump.

#### 3.1.2.1. Column design.

The column design for physical absorption is similar to that used in chemical absorption, but less complicated. For example, the flooding

velocity and the pressure drop are required to design the packed column diameter, and mass transfer coefficient of both the gas and liquid phases are required to design the packing height. To determine the liquid phase mass transfer coefficient in packed columns, the values of the diffusivity, density, viscosity and surface tension are necessary. In addition, the phase equilibrium of the system is important because the saturated absorption amount of the solute in the solvent depends on the vapor-liquid equilibrium of the system.

#### 3.1.2.2. Flash regeneration.

The CO<sub>2</sub> loaded solvent can be regenerated with three different methods: flash at reduced pressure, stripping, and distillation. The process used depends on the required purity. Because the design of the desorber column is similar to the design of the absorber column, identical properties are required. For the flash regeneration, the rich solvent is flashed through several stages to reduce the solubility at a lower partial pressure. Therefore, the phase equilibrium of the mixture is important.

#### 3.1.2.3. Operation.

The operation of the physical absorption process occurs at a very low temperature, particularly in the Rectisol process. The main energy demands of this process are made by the refrigeration system, including the feed gas and the solvent transportation. The required heat transfer area and the cooling load are determined by the heat transfer coefficient, which is related to the thermal conductivity, the heat capacity, the density, and the viscosity of the fluids in the heat exchanger. For shell and tube heat exchangers, the tube-side heat transfer coefficient is a function of the Reynolds number, the Prandtl number, and the tube diameter. The Reynolds number and the Prandtl number can be unpacked into the parameters of density, viscosity, thermal

conductivity, heat capacity, and superficial velocity of the stream.

#### 4. CO<sub>2</sub> capture by physical adsorption

##### 4.1. Process description and related properties of physical adsorption

###### 4.1.1. Process description of physical adsorption

Physical adsorption is another technology for facilitating CO<sub>2</sub> capture from flue gas using a solid adsorbent. Pressure/vacuum swing adsorption (PSA/VSA) and temperature swing adsorption (TSA) are the two main feasible adsorption technologies for large-scale CO<sub>2</sub> separation and purification. In a typical adsorption process, as shown in Fig. 4, the flue gas is compressed and sent to the adsorption column. The CO<sub>2</sub> in the gas stream is captured by the adsorbent, and the impurities are expelled from the top of the adsorption tower. After the adsorption process, the CO<sub>2</sub> is separated, and the adsorbent is regenerated in the desorption tower. The regenerated adsorbent is reused, and the CO<sub>2</sub> is recovered and undergoes further processing. The typical adsorption system consists of the following main components: the compressor, the cooler, the adsorption column, and the desorption column. The compressor is an important component because it demands the most energy in the adsorption process and will be elaborated in the paper when discussing CO<sub>2</sub> conditioning. The adsorption column is also a significant component of the process, and its design and operation greatly impacts the performance of the adsorption rate and the purity of the CO<sub>2</sub> product.

###### 4.1.2. Key components and related properties of physical adsorption and existing studies

In most adsorption processes, the adsorption of the fluid on the adsorbent occurs in a fixed bed. An accurate mathematical model for predicting the performance of the adsorptive capture of CO<sub>2</sub> is necessary for process design and optimization. A comprehensive model consisting of coupled partial differential equations of mass, energy and

the momentum balance with equilibrium equations is necessary. Different properties are required. Specifically, in the mass balance of the adsorption process, the gas–solid adsorption equilibrium is the most important parameter; it controls the dynamic behavior and the adsorption capacity of a packed column. This parameter also determines the mass transfer coefficient. According to the existing models, the mass transfer coefficient is a function of the Sherwood number, which is further related to the Reynolds number and the Schmidt number. Therefore, the properties of CO<sub>2</sub> mixtures, such as their density, viscosity, and diffusivity, are required for determining the mass balance of the adsorption column. In determining the energy balance in the gas phase, it should be noted that the energy flow consists of a heat transfer between CO<sub>2</sub> mixtures and the surface of the solid adsorbent as well as the energy transferred to the column wall. The energy balance can be expressed by an energy conservation equation and is related to the heat transfer coefficient, the bulk density of the CO<sub>2</sub> mixtures, and the heat dispersion. To calculate the heat transfer coefficient in the adsorption column, the properties of CO<sub>2</sub> mixtures such as their density, viscosity, thermal conductivity, and heat capacity are necessary. In determining momentum balance. The pressure drop of the CO<sub>2</sub> mixtures due to viscous energy losses can also be estimated. The pressure drop could reduce the working capacity and is also required for performing the compression work. The pressure drop of the structures in the column can be expressed by the viscosity, density, velocity and properties of adsorbents.

#### 5. Discussion and conclusion

The involved CCS processes and properties (including thermodynamic properties and transport properties) are listed in Table 7. The relationship between the processes and the properties, as well as the status of the property impacts in existing studies are also identified in the Table. Symbol ‘p’ indicates that the property



has been studied and quantitative results are available. Symbol ‘\*’ indicates that no qualitative results. Symbol ‘—’ shows that there is no existing research on the impact of the property. Symbol ‘’ denotes that the property is not related to the process.

For chemical absorption, the impacts of the properties on the process parameters were studied quantitatively. In the case of amine absorption in a packed column, deviation of the density and the heat capacity of the liquid phase were found to have notable impacts on the location and magnitude of the temperature bulge in the liquid phase temperature profile inside the absorber. However, the deviation of the density and the heat capacity causes less significant impacts on the rich stream loading and CO<sub>2</sub> recovery rate compared with the impacts from viscosity and diffusivity. Apart from the liquid phase, the impacts of the gas phase properties, including density, viscosity and diffusivity, were also evaluated. The deviation of gas phase diffusivity was found to have a more significant impact on the packing height design than the density and viscosity. Moreover, a study was found regarding the impacts of the phase equilibrium used to retrieve the activity coefficients of the species for the calculation of the heat of absorption. Phase equilibrium deviation results in different values of the heat of absorption of CO<sub>2</sub> in the amine solution and will directly affect the determination of the energy requirement for the solvent regeneration. Knowledge gaps on the quantitative studies of the property impacts on chemical absorption processes are still required for both transport and thermodynamic properties. There are several parts of the design and operation that have not been studied concerning the property impacts, such as column diameter and pressure drop, especially in the desorption part, which is more complicated than the absorption. Moreover, studies on the impacts of several properties, such as surface tension and thermal conductivity, were not found.

Few existing studies on the property impacts for the physical absorption process were found. Only

the impacts of liquid phase density, heat capacity, enthalpy and phase equilibrium on the Rectisol process were studied. The evaluated process parameters are the temperature, pressure, mole flow and mole fraction of each component of the streams leaving the absorbers. The studies focused mainly on the accuracy of different property models, and the quantitative results of the property impacts are not available. Moreover, the impacts of the transport properties and the impacts on the other parts of the process, such as the refrigeration system and flash regeneration, were not found. Therefore, in this capture process, quantitative evaluations of the property impact on the process parameters are required for both thermodynamic and transport properties.

For the CO<sub>2</sub> adsorption process, there are no studies on the property impacts on the process. However, the properties of CO<sub>2</sub> mixtures, including density, viscosity, heat capacity, phase behavior, thermal conductivity, and diffusivity, are necessary for calculating the momentum, mass and energy balance for evaluation of the performance of adsorption columns.

The evaluation of the performance of membrane separation is dependent on the knowledge of several properties. However, as listed in Table 7, the study of the property impact on the process parameters, such as the required area of membrane and the compression duty, is still lacking.

For CO<sub>2</sub> compression and pumping processes, the CO<sub>2</sub> pumping process is more sensitive to the uncertainty of the properties. Among the properties, heat capacity, density, enthalpy and entropy are the most important ones, and it has been shown that 2% deviation in these properties can result in up to 12% difference in the calculation of the pumping efficiency. Compared with the pumping process, the compression performance is less sensitive to the properties and impurities. The compression performance is most sensitive to heat capacity and compressibility. The impacts of thermal conductivity, speed of sound, viscosity and density still must be evaluated.

However, in some cases, the impacts of different properties were evaluated in one simulation, which gives the synergetic effect of property groups, for example, the study of [112] on thermal conductivity, speed of sound, viscosity and density. Therefore, further research should be performed to investigate the impacts of single property to rank the properties by importance.

For the property impacts on condensation and liquefaction, the impact of density, enthalpy and entropy were studied quantitatively. The deviations of density, enthalpy and entropy using different models are low and give small deviations in the determination of the exhaust gas enthalpy drop in heat exchangers. However, for other related properties, such as phase characteristic, heat capacity, viscosity and thermal conductivity, the impacts must be further investigated.

For the transport process, existing studies mainly focused on property impacts on the performance of pipeline steady flow processes. The studied parameters include pressure loss, temperature drop and transport safety. Among the properties, density and heat capacity are the most important in the pipeline steady flow process. For other properties, including viscosity and thermal conductivity, the property impacts are much smaller. In addition, the phase characteristics were studied mainly from the aspect of avoiding two-phase flow and securing transport safety; these phase characteristics are also very important in pipeline transport. However, the research involving the property impacts on transient pipeline transport processes is insufficient, and the properties, such as speed of sound, diffusivity and Joule–Thomson inversion curves, must be evaluated in the future. In addition, there are no studies conducted on the property impacts on ship transport, which is also a knowledge gap.

In the storage process, the density and viscosity have received most of the attention in the property impact studies, and they were regarded as most important properties concerning the storage capacity and enhanced oil recovery rate. In the

storage capacity calculation, the existence of 15% impurities can change the density and viscosity values up to 50%, which leads to 60% difference in the normalized storage capacity. In addition, the density and viscosity have larger impacts on the EOR process, and 1% change in the density and viscosity can lead to 9% and 6% variation in oil recovery factor, respectively. However, the impacts of the properties of phase characteristic, heat capacity, enthalpy and entropy, diffusivity and thermal conductivity require further studies in the next step in the dispersion process in the EOR process and in the dissolution process in the ocean or the saline aquifer.

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