ORIGINAL PAPER - PRODUCTION ENGINEERING



Diffusion of carbon dioxide in formation water as a result of CO₂ enhanced oil recovery and CO₂ sequestration

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Abstract Flooding carbon dioxide into oil reservoirs is a promising technique for improving the pressure of a reservoir when it is depleted through primary and secondary production. In the context of global warming, it is a viable method for geological storage of CO₂ emissions. Once CO₂ is injected into a reservoir, it is forced to come into partial contact with formation water. To estimate the rate of CO₂ transfer and the total amount of CO₂ dissolved in the formation water, correct estimation of CO2 diffusivity is required. In this study, the rate of CO₂ diffusion in water was experimentally determined in a PVT cell using the pressure depletion method at reservoir conditions (temperature: 50-75 °C and pressure: 17,450 kPa). As expected, the rate of CO₂ diffusion in water increases with increasing temperatures. In addition, the impact of salinity of the water on the rate of CO₂ diffusion was investigated. A significant decrease in the rate of CO₂ diffusion was found with increasing salinity. Subsequently, a diffusion model describing the experiments was developed to predict the behavior of CO₂ diffusivity under simulated conditions. Unique correlations between CO₂ diffusion coefficients and water at different temperatures and salinities were obtained using the results of modeling.

Keywords CO₂ diffusivity · Enhanced Oil Recovery · CO₂ flooding

List of symbols

- \boldsymbol{A} Cross-sectional area of the PVT cell (m²)
- CO₂ concentration in water (kg/m³) \boldsymbol{C}
- Equilibrium concentration of CO₂ in water (kg/m³)
- DDiffusion coefficient (m²/s)
- HHenry's law constant (kPa m³/kg)
- L Height of water in diffusion cell (m)
- M Molecular mass of CO2 (kg/kmol)
- Mass of CO₂ diffused in water (kg) m
- P Pressure (kPa)
- R Universal gas constant
- t Time (s)
- TTemperature (K or C)
- VVolume of CO_2 (m³)
- ZCompressibility factor
- z Depth (m)

Subscript

Initial value

Introduction

Climate change, primarily due to CO₂ emissions, is one of the most important issues facing mankind in this century. Carbon capture and storage (CCS) is currently the most feasible option to reduce billions of tons of CO₂ emissions while offering the continued use of inexpensive, abundant fossil fuels. Storage of CO2 in geological sinks (i.e.,

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depleted gas/oil reservoirs) in conjunction with enhanced oil recovery (EOR) is a potentially attractive technology to limit the accumulation of carbon dioxide in the atmosphere and increase oil recovery.

Recently, CO₂-enhanced oil recovery has been the subject of numerous studies in the petroleum industry (Godec et al. 2013; Ravagnani et al. 2009; Saeedi et al. 2011). Injecting CO₂ under supercritical conditions favors miscibility, decreases the viscosity of the oil in the reservoir and facilitates displacement. In cases where the pressure of the reservoir is depleted through primary and secondary production, water is injected to restore the pressure within the formation to a suitable pressure for CO₂ flooding. Once the CO₂ is injected into the formation, it interacts with oil and water in the pores and over months to years creates a region in which oil saturation and mobility are increased. The amount of CO2 that enters into oil and water is controlled by the rate of CO₂ diffusion in each of them. Analytical and geocellular flow models which are used to estimate the rate of CO₂ injection in CO₂ flooding processes should consider the amount of CO₂ wasted in the water. This requires the correct estimation of CO₂ diffusivity and solubility in the formation water under practical reservoir conditions.

It is well known that the rate of CO2 diffusion into formation water is highly dependent on the reservoir conditions (i.e., pressure, temperature). There is a significant body of relevant literature regarding the effect of pressure and temperature on the rate of CO₂ diffusion in water (Hirai et al. 1997; Leaist 1987; Mazarei and Sandall 1980; Tan and Thorpe 1992). Tamimi et al. (1994) studied the diffusivity of several gases, including CO2, into water at a temperature range of 20-95 °C and at atmospheric pressure. Jähne et al. (1987) measured the rate of CO₂ diffusion in both pure water and sea water using the modified Barrer method. Their experiments were carried out in the temperature range of 5-35 °C under fairly low pressures. Mutoru et al. (2011), who conducted a literature survey on experimental measurements of diffusion coefficients of CO₂-H₂O mixtures, concluded that most of the experimental data on diffusion coefficients of CO2 in water are limited to atmospheric pressure at the temperature range of 0-95 °C and that only a few studies (Hayduk and Minhas 1982; Siddigi and Lucas 1986; Nakanishi 1978) have been performed at pressures close to reservoir conditions.

Salinity of water is another factor affecting the diffusivity of CO₂ in formation water. Salinity of formation water can change over time as the water and rock interact and as reservoir fluids are produced and replaced with water from other formations, injected water or other injected fluids. King et al. (1995), who studied the diffusivity of different gases in pure and sea water, reported that there is an uncertainty associated with estimating the molecular diffusion coefficient of a gas in

seawater from the values obtained in pure water, demonstrating the poor understanding of the effect of salinity on diffusion. The rate of CO_2 diffusion in saline water has been reported by only a few researchers at certain pressures and temperatures (Nazari Moghaddam et al. 2012; Bahar and Liu 2008; Sell et al. 2012). However, as the experimental data are so limited, many aspects of the effect of salinity on the rate of CO_2 diffusion are not well understood; further clarification is required to better understand the role of salinity on the rate of CO_2 diffusion.

In this paper, the results of a systematic experimental investigation on the effect of temperature and water salinity on the rate of CO₂ diffusion in formation water at reservoir conditions are presented. A diffusion model is developed, based on Fick's second law of diffusion and a mass balance between two phases (CO₂–water), to predict the rate of CO₂ diffusion during pressure decay experiments at different operating conditions.

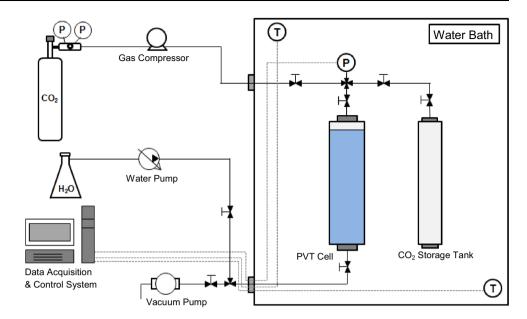
Experimental

The materials used in this work were CO₂ with a purity of 99.99 % and a sodium chloride solution representing the formation water. The sodium chloride solution was made by dissolving pure sodium chloride in distilled water.

Figure 1 shows a schematic diagram of the experimental setup; it was used to measure the mass transfer rates of CO₂ in water inside a closed PVT cell with a diameter of 5.5 cm and length of 28.5 cm at a constant temperature. The entire PVT cell assembly was placed inside a water bath to maintain a uniform temperature throughout the cell. Two thermocouples located above and below the cell were used to monitor the cell temperature. A high-accuracy pressure transducer (± 0.05 % full scale) with the pressure range of 3000 psia was connected to the top of the PVT cell to measure the pressure of the PVT cell during the experiment. Since the density of CO₂ in the experimental conditions is lower than the water, the top port of the PVT cell was connected to a gas storage cylinder with a volume of 365 cm³ through a needle valve, while the bottom port was connected to a water supply container. The feed gas (i.e., CO₂) was supplied to the gas storage cell by a pressurized cylinder and flowed through a high-pressure gas compressor to increase the pressure inside the gas storage cell. The water sample was introduced to the PVT cell by using a high-performance piston pump with a set point of <2%accuracy. The bottom port was also connected to a vacuum pump for evacuating the PVT cell before the experiment. To insure the connections and cells involved in the setup were free of leakage, the entire system was pressurized by CO₂ and the pressure of the system was monitored to detect any possible leakage for 24 h.



Fig. 1 Schematic diagram of experimental setup



A typical test is initiated by heating the entire system to a pre-determined temperature, while it is placed under vacuum to completely remove unwanted gases inside the cells. Once a stable condition is reached, a certain volume of formation water (i.e., 650 cm³) is injected into the bottom of the PVT cell. Simultaneously, the gas storage cell is filled with CO₂ to reach a pre-determined pressure. The whole setup is maintained at the experimental temperature for 24 h before starting the diffusion test. The test is started by opening the valve between the PVT cell and the gas storage cell. As the experiment progresses, the pressure of the system is recorded to calculate the mass transfer rates of CO₂ in the water.

Results and discussion

As described in the "Experimental" section, the CO_2 mass transfer rates were estimated based on the measurement of CO_2 pressure in a PVT cell that contained a column of formation water below a column of CO_2 . Since the temperature of the system was constant during the experiments, the changes in the CO_2 pressure were assumed to be entirely due to the transfer of CO_2 into the water phase. Figure 2 presents a series of three pressure decay tests and their temperature profiles during the experiment. The experiments were conducted at temperature of 68 °C with pure water. The results confirm that the diffusion tests can be reproduced with a high degree of accuracy (± 0.2 %).

The results shown in Fig. 2 also indicate that the CO_2 mass transfer rate, which is proportional to the rate of pressure decline, is significantly higher at the initial times

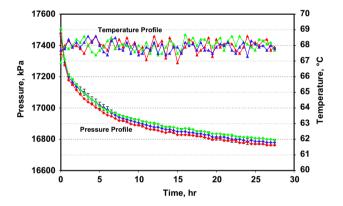


Fig. 2 Reproducibility of pressure curves

of the experiment, gradually reducing to a constant value at longer times (>20 h). These experimental behaviors are consistent with those presented by Farajzadeh et al. (2007); their report showed that the CO₂ mass transfer rate in water is significantly higher at the early steps of the experiment. Similarly, Yang and Gu (2006), who studied the mass transfer of CO2 in a high-pressure PVT cell, suggest that the higher mass transfer rate of CO2 into water at early times is explained by the sharp concentration gradient of CO₂ in the water phase; this results in a density gradient in the water column and promotes the natural convection flow in the region. The effect of density gradient on the rate of CO₂ diffusion in pressure decay experiments has been addressed adequately in the literature. Gholami et al. (2015) studied the effect of natural convection in water-CO₂ system where they conducted pressure decay experiments with permeable glass beads to control convective mixing.



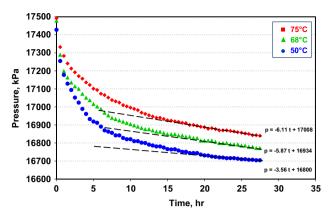


Fig. 3 Effect of temperature on the rate of CO₂ dissolution

Effect of temperature

The effect of temperature on the rate of CO₂ mass transfer in pure water with the initial pressure of 17,450 kPa is shown in Fig. 3. The results indicate that at higher temperatures, the transition from the initial convection regime to the slow diffusion regime takes place at higher pressures, which can be attributed to lower CO₂ solubility at higher temperatures. Furthermore, by comparing the slopes of the pressure curves at different temperatures in Fig. 3 and knowing that the effect of convection on the rate of CO₂ mass transfer is negligible at longer times (>20 h), it can be concluded that the rate of CO₂ diffusion slightly increases with increasing temperatures. The positive effect of temperature on the rate of CO₂ diffusion can be attributed to the increase in the fluctuations of CO₂ molecules as well as the decrease in the viscosity of water at higher temperature.

Effect of salinity

To investigate the effect of formation water salinity on the rate of CO_2 dissolution, a series of tests were conducted with pure water containing different concentrations of NaCl salt (i.e., 20, 40 and 80 parts per thousand) at 68 °C. The results, presented in Fig. 4, clearly indicate that the addition of salt, even at the low salinity of 20 parts per thousand, significantly reduces the overall solubility of CO_2 in water. The results shown in Fig. 4 also suggest that the presence of salt (i.e., NaCl) in the solution reduces the rate of CO_2 mass transfer in the longer steps, where the overall process is controlled by the diffusion between CO_2 and water. Similar behavior is also reported by Jafari Raad et al. (2015) where they found that increasing ion concentration in the water solution decreases diffusivity of CO_2 due to growth in fraction of contact ion pairs. To

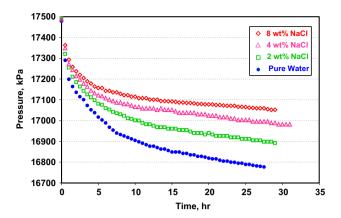


Fig. 4 Effect of salinity on the rate of CO₂ dissolution

explain this behavior, it can be argued that by dissolving NaCl in water, the ionic strength of the solution increases; this results in higher electrostatic attraction between the layers of water, thus increasing the viscosity of the water. As the water viscosity increases, the CO₂ molecules move more slowly around water layers. It is clear that the diffusion is directly related to the motion of molecules in a fluid; therefore, it is logical to assume that the rate of CO₂ diffusion decreases as the viscosity of water increases. The effect of liquid viscosity on the rate of CO₂ diffusion is well understood in the literature. Several published studies related the rate of CO₂ diffusion in liquids over a broad range of liquid viscosities, providing excellent correlation for all experimental data in the literature (Hayduk and Cheng 1971; McManamey and Woollen 1973).

Physical model

The experimental data obtained in this study were used to develop a diffusion model to predict the rate of CO_2 diffusion in various operating conditions. The model describes the rate of diffusion using Fick's second law of diffusion coupled with a mass balance at the interface of CO_2 and water. Due to its simplicity, this model has been commonly used in the literature to determine the diffusivity of gases in liquids (Riazi 1996; Zhang et al. 2000; Sheikha et al. 2005). A schematic diagram of the diffusion model is shown in Fig. 5, where the interface between two phases is located at z=0 and the bottom of the PVT cell is located at z=L. The diffusion process is governed by the unsteady one-dimensional diffusion equation as:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial z^2} \tag{1}$$

where C is the CO₂ concentration in water; z is the distance from the top of the water phase; t is time; and D is the



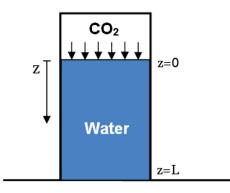


Fig. 5 Schematic of the CO_2 -water model inside a closed diffusion cell

diffusion coefficient of the CO₂ in water, assuming that it is constant throughout the diffusion process.

The assumption of a constant diffusion coefficient in the CO_2 -water is reasonable, because the CO_2 concentration in water is generally low under the test conditions. For the same reason, the swelling effect is negligible. The model also assumes that the density in the CO_2 phase only depends on time and that the CO_2 concentration at the water- CO_2 surface is estimated by the thermodynamic equilibrium of water and CO_2 , as described by Henry's law.

At the beginning of the diffusion test, the water is free of CO₂. Thus, the initial condition (IC) is given by:

$$C(z,0) = 0 \quad 0 < z < L \tag{2}$$

For the impermeable rigid boundary at the bottom of the PVT cell, given that the diffusion process is slow and the water column is long, it is assumed that the concentration of CO_2 is zero at the end of the cell $(L \to \infty)$. Later, it will be shown that the infinite-acting boundary condition is a valid assumption under the test conditions.

$$C(L,t) = 0 (3)$$

The concentration of CO₂ in the water at the interface, where the two phases are at equilibrium, can be expressed in terms of pressure using Henry's law as follows:

$$P = HC(0, t) \tag{4}$$

According to the interface boundary condition, the CO_2 mass transfer flux across the interface is proportional to the difference between the existing mass of CO_2 and the initial mass of CO_2 in the CO_2 storage cell. The mass transfer rate of CO_2 diffused at the interface can also be calculated from Fick's first law of diffusion. The corresponding mass balance is given as:

$$\frac{\mathrm{d}m}{\mathrm{d}t}\Big|_{z=0} = \frac{VM}{ZRT} \frac{\mathrm{d}P}{\mathrm{d}t} = DA \frac{\mathrm{d}C}{\mathrm{d}z}\Big|_{z=0}$$
 (5)

where V and A are the CO_2 volume and the cross-sectional area of the cell, respectively, R is the universal gas constant

and Z is the compressibility factor. By substituting Henry's law for the CO_2 pressure in Eq. (3), the interface boundary condition can be written in the following form:

$$\frac{\mathrm{d}C}{\mathrm{d}t}\Big|_{z=0} = \frac{DAZRT}{VMH} \frac{\mathrm{d}C}{\mathrm{d}z}\Big|_{z=0} \tag{6}$$

The mathematical formulation, given by Eqs. (1) and (6) was solved using the Laplace transform method. The analytical solution for the above mass transfer problem was obtained by Riazi (1996) as:

$$C(z,t) = \frac{P_{i}}{H} \exp\left(\frac{z}{\eta D}\right) \exp\left(\frac{t}{\eta^{2} D}\right) erfc\left(\frac{\sqrt{t}}{\eta \sqrt{D}} + \frac{z}{2\sqrt{Dt}}\right)$$
(7)

where $\eta = \frac{VMH}{\sqrt{D}AZRT}$. From the above concentration distribution, the pressure of CO₂ in the CO₂ storage tank is found to be:

$$P(t) = P_{i} \exp\left(\frac{t}{\eta^{2}}\right) erfc\left(\frac{\sqrt{t}}{\eta}\right)$$
 (8)

In Eq. (8) the only parameter of the model (D) is determined by the approximation suggested by Sheikha et al. (2005) (Eq. 9), where the exponential term in Eq. (8) is ignored as it is close to one at the range of predicted values for D.

$$\frac{\mathrm{d}[erfc^{-1}\{P(t)/P_{\mathrm{i}}\}]}{\mathrm{d}(\sqrt{t})} = \frac{\sqrt{D}AZRT}{VMH}.$$
(9)

In Eq. (9), independent evaluation of Henry's constant is necessary to determine the diffusion coefficient. In the present study, Henry's constant was calculated using a correlation for the solubility of CO₂ in water obtained by Duan and Sun (2003). The solubility was converted to an average value of Henry's constant, using the following relationship:

$$H = \frac{P}{C_{\text{eq}}}. (10)$$

With the calculated value of Henry's constant, the diffusivity coefficient is determined using Eq. (9).

Figure 6 shows a typical plot of $erfc^{-1}\{P(t)/P_i\}$ versus the square root of time; it indicates that the initial slope of the curve is higher than at later points, which is explained by the impact of natural convection during the early steps. The results also indicate that the slope of the curve becomes linear at longer times, suggesting that the mass transfer of CO_2 into water is controlled by classical molecular diffusion.

The diffusion coefficient of CO_2 was calculated using a linear correlation—fitted to the curve in Fig. 6 at both convection-controlled and diffusion-controlled regions—together with Eq. (9). The results indicate that the diffusion



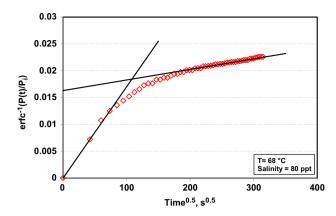


Fig. 6 Profile of $erfc^{-1}\{P(t)/P_i\}$ versus the square root of time during the experiment

coefficient obtained in early stages of the experiment $(1 \times 10^{-7} \text{ m}^2/\text{s})$ is two orders of magnitude higher than the one obtained at longer times $(3.6 \times 10^{-9} \text{ m}^2/\text{s})$ which again confirms the effect of convection at the early stages of the experiment. Clearly, the diffusion coefficient obtained at the convection-controlled region takes into account the phenomenon of natural convection and fails to predict the correct diffusion coefficient of the system. A similar conclusion has been reported by Gholami et al. (2015) where they concluded that the application of the inverse methods to find diffusivity from the entire pressure—time data leads to significant errors.

The effects of temperature and salinity on the diffusivity of CO₂, extracted from the behavior of the experiments at longer times, are shown in Figs. 7 and 8. The results clearly indicate that the diffusion coefficient of CO₂ in formation water increases with increasing temperatures and decreases with increases in the salinity of formation water. The results obtained in Figs. 7 and 8 are in good agreement

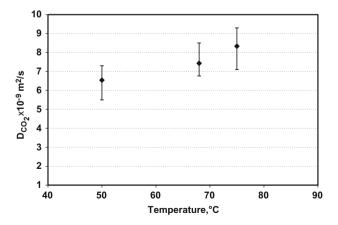


Fig. 7 Effect of temperature on diffusion coefficients of CO₂ in pure water

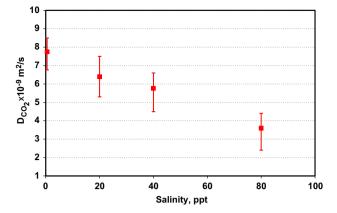


Fig. 8 Effect of salinity on diffusion coefficients of CO_2 in formation water ($T=68~^{\circ}C$)

with the diffusivity measurements in the literature at high pressures. Lu et al. (2013) studied the diffusivity of carbon dioxide in water in a high-pressure capillary optical cell. They reported that the diffusion coefficient of carbon dioxide improves from 3.2×10^{-9} to 5.3×10^{-9} m²/s with an increase in temperature from 50 to 80 °C at the constant pressure of 20 MPa. Renner (1988) also found that the diffusion coefficients of CO₂ in consolidated porous media saturated with NaCl change in the range of 3×10^{-9} – 7×10^{-9} m²/s when pressure increases from 1544 to 5833 kPa at a constant temperature of 38 °C. Furthermore, Azin et al. (2013) employed the pressure decay method to measure the diffusivity of CO₂ in a saline aquifer (115 ppt NaCl) and reported that the diffusivity of CO_2 varies in the range of 3.52-6.16 \times 10⁻⁹ m²/s at the pressure range of 5900-6900 kPa and the temperature range of 32-50 °C.

The performance of the proposed model was also compared against the Mutoru–Dios–Firoozabadi model (Mutoru et al. 2011) which is capable of predicting diffusion coefficients for carbon dioxide and water mixtures at both low and high pressures. This model takes into account the temperature effect on the total dipole moment of water and the induced dipole moment on CO_2 to accurately calculate the diffusion coefficient of CO_2 in water. The model prediction in the temperature range of 50–75 °C and pressure of 17 MPa varies from 3.9×10^{-9} to 6.1×10^{-9} m²/s. Thus, the model proposed in the current study performs fairly well when compared to other models in the literature.

To accurately calculate the concentration profile of CO_2 along the cell, the diffusion equation (Eq. 1) with molecular diffusivity has to be solved simultaneously with the Navier–Stokes equation to consider the effect of natural convection on the CO_2 profile in the PVT cell. As this becomes a complicated numerical problem, a diffusion



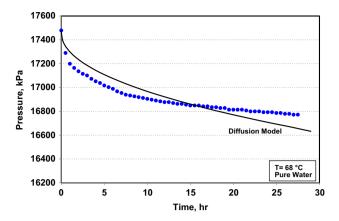


Fig. 9 Comparison of model and experimental data

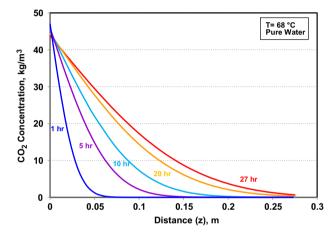


Fig. 10 CO₂ concentration in the water phase

equation (Eq. 1) with an effective diffusion coefficient was used in this study to obtain the profile of CO₂ along the cell. The effective diffusion coefficient was determined by finding the minimum average pressure difference between the model and experimentally measured pressures at different times. Figure 9 presents the comparison between the measured pressures and the predictions of the model using the effective diffusion coefficient of CO₂ in water. Figure 10 shows the concentration profile of CO₂ along the PVT cell, confirming that the infinite-acting boundary condition is a valid assumption under the test conditions.

Conclusion

Diffusivity of CO₂ in formation water was experimentally studied at reservoir temperatures and high pressures (i.e., 17,500 kPa) using the pressure decay method in a PVT cell. The results obtained in this study show that the rate of CO₂ diffusion in water was much higher at the early stages of the

experiments. This is probably due to natural convection caused by the density gradient in the water column resulting from CO_2 dissolution. The results also indicate that the rate of CO_2 diffusion in formation water increases with increasing temperatures at the range of 50–75 °C, which is due to the fact that CO_2 molecules move faster at higher temperatures. Furthermore, the experimental results demonstrate that the rate of CO_2 diffusion decreases significantly with increasing NaCl concentration in formation water. The presence of salt in formation water is believed to increase the viscosity of the water, which results in higher resistance for the movements of CO_2 molecules through water layers.

A simple diffusion model based on Fick's second law and Henry's law was employed to estimate the diffusion coefficients of CO_2 at the temperature and salinity range studied. The values obtained at the later stages of the experiments were considered as diffusion coefficients of CO_2 in formation water; they agreed with the reported data in the literature for molecular diffusivity of CO_2 in water.

It would be informative to expand the experimental conditions in this study, repeating the experiments at higher pressures and temperatures to see if similar behavior is observed. It would also be useful to study the effects of other common salts in formation water on rates of CO₂ diffusion.

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