IS CARBON DIOXIDE AN ORE-FORMING FLUID UNDER SHALLOW EARTH CONDITIONS?

By R. M. Garrels and D. H. Richter

Trace Elements Investigations Report 472

UNITED STATES DEPARTMENT OF THE INTERIOR

GEOLOGICAL SURVEY
Dr. T. H. Johnson, Director
Division of Research
U. S. Atomic Energy Commission
16th Street and Constitution Avenue, N. W.
Washington 25, D. C.

Dear Dr. Johnson:

Transmitted herewith is one copy of TEI-472, "Is carbon
dioxide an ore-forming fluid under shallow earth conditions?" by

We plan to submit this report for publication in Economic
Geology.

Sincerely yours,

[Signature]
for W. H. Bradley
Chief Geologist
IS CARBON DIOXIDE AN ORE-FORMING FLUID UNDER
SHALLOW EARTH CONDITIONS?*

By

R. M. Garrels and D. H. Richter

December 1954

Trace Elements Investigations Report 472

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IS CARBON DIOXIDE AN ORE-FORMING FLUID UNDER SHALLOW EARTH CONDITIONS?

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ABSTRACT

A review of some of the physical chemical properties of \( \text{CO}_2 \) and of the system \( \text{CO}_2-\text{H}_2\text{O} \) indicates that under some shallow earth conditions \( \text{CO}_2 \) can exist as a separate phase with a density approximately that of water. The effect of dissolved neutral or acid salts on the solubility of \( \text{CO}_2 \) in \( \text{H}_2\text{O} \) is negligible. Carbonates, oxides, and silicates, through reactions with a \( \text{CO}_2 \)-saturated water phase, will eventually cause the disappearance of a \( \text{CO}_2 \) phase. The prevalence of \( \text{CO}_2 \) in the earth's crust is attested by its abundance in natural gases and fluid inclusions in minerals.

Some of the characteristics of uranium ores of the Colorado Plateau are compatible with the assumption that deposition was from a fluid \( \text{CO}_2 \) phase.

INTRODUCTION

During the course of some speculations regarding the origin of the sandstone-type uranium deposits, we were led to consider the possibility that a non-aqueous phase might be the agent of metal transport and deposition. Because of the prevalence of carbon dioxide in earth materials of all kinds, a review of the properties of carbon dioxide under shallow earth conditions seemed warranted. Although we do not have the temerity to advocate carbon dioxide as
the ore-forming medium in such deposits, the results of the review were so opposed to our preconceived ideas concerning the nature of this gas that we are impelled to present them for general consumption.

Among the questions that occurred to us at the start of the investigation were:

Do water and carbon dioxide exist as separate phases in rocks? If so, what is the concentration of water in the carbon dioxide phase, and vice versa? What is the density of carbon dioxide under shallow earth conditions? What is known about the solvent properties of carbon dioxide?

PART I - THE SOLID PHASE

P-V-T relations for CO₂

The pressure-volume-temperature relations for CO₂ are shown in figure 1 (Michels and others, 1935, 1935a; and Kennedy, 1954). Superimposed on the diagram are lines showing high, "normal", and low thermal gradients under hydrostatic pressure conditions, and the same gradients for lithostatic conditions. As an example, let us briefly examine some of the properties of CO₂ along a "normal" thermal gradient (1°C/100 ft.) under lithostatic pressures as shown on the P-V-T diagram. At a temperature of 110°C and pressure of 800 atmospheres, corresponding to a depth of 10,000 feet, CO₂ has a specific volume of 1.08, or in other words, a density of 0.93. At decreasing temperatures and pressures, the CO₂ volume slowly increases, concomitant with a slow decrease in density, until near surface conditions are reached. Here at 15°C and 50 atmospheres (750 feet below the
Figure 1. -- P-V-T diagram for CO$_2$ with superimposed high, "normal", and low thermal gradients under lithostatic and hydrostatic pressures. Specific volumes (sloping solid lines) are expressed in cc/g. Greatest density is reached under a high pressure-low temperature gradient (left hand dashed line), least density under hydrostatic pressure and a high temperature gradient (right hand dashed line).
surface), equivalent to the intersection of the earth gradient and CO₂ vapor pressure curve, CO₂ rapidly expands filling a volume eight times that of its original size. With further decrease in temperature and pressure CO₂ continues to expand rapidly until equilibrium is established with atmospheric conditions. The lithostatic pressure gradient must represent roughly the maximum that can exist for appreciable time intervals. Gas or fluid pressures, built up from below, are greater than the hydrostatic pressure by the amount necessary to overcome the frictional resistance of the enclosing rocks. In unfractured and slightly permeable rocks the pressure must exceed the rock load if the fluid is to move at an appreciable rate. That such conditions do exist is shown by measured pressures in drill holes; in fact, Pinfold (1954, p. 1660) reports bottom hole pressures in excess of the lithostatic load. The temperature gradients chosen are well within the observed range (Lindgren, 1919, p. 80-84, and Van Orstrand, 1934). The lithostatic pressure is computed on the assumption that the average rock density is 2.7.

CO₂-H₂O solubility relations

The solubility of CO₂ in water has been determined by Wiebe and Gaddy (1939, 1940); the solubility of H₂O in gaseous CO₂ should be roughly proportional to the partial pressure of H₂O. On this basis the weight ratio of water to CO₂, for a given pair of P-T values, is taken as proportional to the ratio of the vapor pressure of water to the total pressure on the system (P_{CO₂} + P_{H₂O}). For actual calculation of the mole percent water in the gaseous CO₂, the Gas Law was used.
Some question may arise as to the validity of the assumption that CO₂ at high density acts as a gaseous solvent, or that water vapor itself under such high total pressure obeys the Gas Law. However, determinations of the solubility of water in liquid CO₂ (Stone, 1943) suggest that serious error in the order of magnitude of the calculated solubilities is not likely. At 22.5°C, and under the vapor pressure of CO₂ (60 atmospheres), the solubility of water in liquid CO₂ is only 0.954 weight percent. Furthermore, the data at temperatures up to 22.5°C indicate that the solubility of water in liquid CO₂ increases with temperature at a decreasing rate under the equilibrium vapor pressure. Because earth gradients have linear increase in pressure, whereas the vapor pressure curve of CO₂ is exponential, it appears very likely that no great error is involved in the calculated solubility.

Figure 2 shows the composition of the water phase and the carbon dioxide phase for a mixture of carbon dioxide and water, transported down a thermal gradient of 1°C per 100 feet under lithostatic conditions. The low mutual solubility is apparent.

Figure 3 shows iso-solubility curves for CO₂ in water on a P-T graph. The nearly constant solubility at low temperatures and pressures along all the gradients chosen, as well as the marked increase at the higher ranges, are noteworthy features. In other words, a saturated solution of CO₂ in water, if it moves upward from depth, will separate a nearly pure fluid CO₂ phase. This deduced behavior is substantiated by observations on certain three-phase fluid inclusions in many minerals. The three phases are two liquids and a gas at room temperature. If the inclusions are heated one of the liquid
Figure 2.--Solubility relations of $\text{CO}_2$-$\text{H}_2\text{O}$ along a "normal" thermal gradient and under a lithostatic load.
Figure 3.--P-T diagram for CO₂ with iso-solubility lines for CO₂ in H₂O. The figures are expressed in cc/gram at STP. (After Wiebe and Gaddy). Also shown are high, "normal", and low thermal gradients under lithostatic and hydrostatic pressures. Note that the solubility relations are such that there is very little change with depth, irrespective of the specific earth gradient. For example, under lithostatic pressure, and under a temperature gradient of 10/150' (left hand dashed line), the solubility is essentially constant from 50 to 500 atmospheres pressure, and increases only from 35 cc CO₂/gram H₂O at 500 atmospheres to 40 cc/gram at 850 atmospheres.
phases disappears at 31°C, the critical temperature of carbon dioxide. The remaining gas and liquid phase commonly persist to temperatures about 200°C; then the gas phase disappears and the inclusions fill with liquid (Cameron and others, 1953). On cooling, the sequence of events is reversed. The disappearance of the vapor phase, on heating, is by no means entirely attributable to the expansion of the liquid phase but must also indicate a high solubility of CO₂ in H₂O at temperatures above 200°C and high pressures. Further evidence of the existence of a nearly pure dense gaseous CO₂ phase under earth conditions is shown by numerous fluid inclusions that exhibit only a gas phase above the critical temperature of CO₂ and a liquid phase with a small gas bubble below the critical temperature. The absence of appreciable water in the CO₂ phase is shown by the disappearance of liquid CO₂ at the critical temperature for pure CO₂. Appreciable water in solution would raise the critical point.

Density relations

To illustrate density changes it is instructive to take a mixture of one mole of CO₂ and one mole of water from the earth's surface down various earth gradients. At the surface the volume discrepancy of the two phases is huge; 18 cc. for the liquid water and 23,100 cc. for the gaseous CO₂, but the descent into the earth changes the picture considerably.

In figure 4 the changes are shown diagrammatically. CO₂ shrinks as pressure is applied and reaches a density approaching that of water at depths of a few thousand feet, depending upon the specific earth gradient. Thus under natural conditions it is probable that at relatively shallow depths a fluid CO₂ phase may exist with a density equal
Figure 4.—Diagrammatic representation of the history of an equimolar mixture of CO$_2$ and H$_2$O down various temperature-pressure gradients. The circles are sections of spheres representing the relative volume of the CO$_2$-rich phase and the H$_2$O-rich phase. The change from a "dense liquid-rare gas" system at the surface to a "dense liquid-dense gas" system takes place at shallow depths under all gradients shown.
to or slightly greater than that of water! In fact, in moderate to high latitudes, where the near-surface temperature is low, liquid CO₂ may form.

In figure 5 the density of CO₂ as a function of depth is shown graphically, to emphasize the high density that is reached at relatively shallow depths.

CO₂, under most earth conditions, has a density greater than that of liquid CO₂ at room temperature. There is no change in solvent properties of a liquid phase as it changes to a gas, if there is no change in density, so dense gaseous CO₂ can be expected to behave like the liquid (cf Hannay and Hogarth, 1879; Ingerson, 1934, pp. 457-458; Smith, 1943, p. 34). The fact that in many environments, it is not only denser, but hotter than the liquid, indicates that it probably is considerably more effective as a solvent. The density of liquid CO₂ at 31°C, under its vapor pressure of 73 atmospheres, is 0.47, whereas at 100° and 1000 atmospheres, the density of the fluid is about 1.0.

Effect of dissolved material on CO₂ solubility in water

So far the discussion has been concerned only with the system CO₂-H₂O. The question arises as to the effect of other dissolved materials on the CO₂ solubility in water. Most dissolved salts reduce gas solubility, if no chemical reaction between the dissolved gas and the solutes occurs (Mellor, 1925, p. 53). Such reaction by dissolved CO₂ with neutral or acid salts is not to be expected; a long list of
Figure 5.--Density of CO₂ under various shallow earth conditions.
determinations of CO$_2$ solubilities in a variety of such salt solutions (Seidell, 1940, pp. 225-230) shows remarkably uniform behavior and a relatively small salt effect.

Dissolved CO$_2$ will react with dissolved basic salts, such as sodium carbonate, but the solubility is probably most affected by the reaction of the CO$_2$-H$_2$O system with rock minerals susceptible to acid attack. If a "bubble" of dense CO$_2$ were injected into a water-saturated sandstone that had a calcite or an iron oxide cement, CO$_2$ would dissolve in the water and then react with the iron oxide and calcite. As a result of the reactions, the bubble would eventually disappear. The reactions would be:

$$\text{CO}_2 + \text{H}_2\text{O} \rightarrow \text{H}_2\text{CO}_3 \rightarrow \text{H}^+ + \text{HCO}_3^-$$

$$\text{CaCO}_3 \downarrow$$

$$\text{Ca}^{++} + \text{HCO}_3^-$$

$$6\text{CO}_2 + 6\text{H}_2\text{O} \rightarrow 6\text{H}_2\text{CO}_3 \rightarrow 6\text{H}^+ + 6\text{HCO}_3^-$$

$$\text{Fe}_2\text{O}_3 \downarrow$$

$$2\text{Fe}^{+++} + 3\text{H}_2\text{O}$$

The same type of reaction would take place with silicate minerals but at a much slower rate.

In general, then, in a low temperature system of CO$_2$ and H$_2$O, a separate CO$_2$ phase will exist even if the weight percent CO$_2$ is small; but, if the system is placed in contact with rocks containing carbonates or oxides, the CO$_2$ phase will gradually disappear as it dissolves into the water to replace that used up by reaction.
A separate CO₂ fluid phase would thus not be expected to persist indefinitely if it were moving through water-saturated rocks, unless they were pure quartz sandstones, or perhaps certain acid resistant clays. Certainly it would tend to disappear in limestones, or from oxide or calcium carbonate cemented sandstones, siltstones, or shales.

The rate of disappearance would be a complex variable, depending on the rate of reaction of the solids and the rate of diffusion of CO₂ into the water phase. The second variable would in turn depend on whether or not the CO₂ was moving relative to the water phase.

The persistence of gaseous CO₂ as a separate phase in nature is shown by its presence in the exhalations of many volcanoes and hot springs, as well as by its occurrence as a major constituent in some natural gases from stratigraphic or structural traps (table 1). Concerning its ubiquitous nature in fluid inclusions in minerals Lindgren (1919, p. 95) eloquently states: "The presence of liquid carbon dioxide in cavities in minerals of igneous rocks is proof of its occurrence in the molten magma consolidated in depth. Every eruption brings new evidence of exhalations from magmas congealing near the surface; and almost every volcanic district of recently closed igneous activity testifies to the persistence of this gas in escaping from the cooling lavas below."

SUMMARY

In summary, CO₂ can exist as a separate phase under shallow earth conditions. At depths below 1,000 feet, under a "normal" temperature gradient and a lithostatic load, its density is equal to that of liquid CO₂ at the critical point. Under high pressure gradients and
Table 1.—The CO₂ content of some earth gases.

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<td>Clarke (1924, p. 269) after Moissan</td>
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<td>Yellowstone National Park, Wyo., average of 40 analyses</td>
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<td>San Rafael swell, Utah</td>
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low temperature gradients its density approximates that of water at depths between one and two miles. Dense fluid CO₂ probably has solvent properties more nearly comparable to a liquid than to those generally attributed to a gas. Unfortunately, little is known of the solvent properties of liquid CO₂. In most rocks a separate CO₂ phase probably would disappear over a long period of time as a result of reaction of the CO₂-saturated water phase with rock minerals susceptible to acid attack.

PART II - THE GASEOUS PHASE

CO₂ and uranium deposits

The relatively common occurrence of fairly pure gaseous CO₂ in nature, plus the fact that its density, even under shallow earth conditions, is greater than that of liquid CO₂ at room temperature, leads naturally to the possibility that CO₂ may play an important role in the transportation and deposition of other compounds. Present knowledge of the solvent properties of dense gaseous CO₂, or even of liquid CO₂, is very limited. and certainly merits extensive investigation.

To show the part that dense CO₂ might play as a transporting agent, it is intriguing to consider to what extent it would fit the characteristics required of the fluid that deposited the uranium ores of the Colorado Plateau. Rather than attempt any coherent reconstruction of the details of the processes of deposition, we prefer to assemble a somewhat disconnected group of arguments, explanations, and generalizations, which we hope will prove provocative, and may lead others to more detailed examination of some of the aspects considered.
First, the best reconstruction of the conditions of deposition of the ores indicates that CO₂ would have an appropriate density. According to Stieff and Stern (1952, p. 708), the ores were deposited in early Tertiary or Late Cretaceous time. At the end of Cretaceous, the ore-bearing Triassic or Jurassic beds were buried by about 10,000 feet of sediment, so that, on the basis of a "normal" geothermal gradient, and a hydrostatic load, the temperature would have been about 110°C, and the pressure about 300 atmospheres. The density of CO₂, under these conditions, is about 0.6, roughly that of liquid CO₂ at room temperature under its equilibrium vapor pressure. If it is considered that the geothermal gradient would probably be a little lower in a basin of sinking sediments, and that the pressure on ascending CO₂, held back by the frictional resistance of the nearly impermeable shales of the Chinle, Brushy Basin, and Mancos formations, might well be lithostatic, the density of CO₂ would be about that of the pore waters of the mineralized sediments. The relation of the ores to the containing beds suggests strongly that the density of the ore fluid was equal to or greater than the pore waters; in the Shinarump conglomerate, ore is characteristically in channels at the base of the formation, whereas in the Salt Wash sandstones of the Morrison formation, it commonly "floats" in the middle of an apparently homogeneous sandstone layer. Elsewhere in the Salt Wash, the ore may "hang" against a shale split for long distances, and then abruptly cut down across bedding planes. In still other places it hugs the base of a sandy layer.

The "bleached" shales that so often underlie or overlie ore bodies might be the result of the slow solution of CO₂ in the pore waters of the rock, with concomitant removal of hematite cement and solution and
recrystallization of calcite. The effect of CO₂ dissolving in pore waters might also account for the occurrence of ore in light-colored sandstones, often with disseminated pyrite. The CO₂ dissolved in the pore waters would react with the iron oxide of originally red sediments, and the resulting ferric iron would be reduced and precipitated as pyrite by a small amount of H₂S in the CO₂, or could even be adsorbed by, or taken up in the structure of clay minerals (Gruner, 1953, p. 28-34).

The rhythmic precipitation that is so common in the ores, and has led some workers (Fischer, 1942, p. 389) to postulate a groundwater origin, might be rhythmic precipitation at the shrinking boundary of the dissolving CO₂ phase.

The very sharp, sometimes undulating margins of many ores, suggest deposition at a phase boundary, perhaps between pore waters and the dense gaseous CO₂.

The fact that the ores represent additions of an unusual suite of elements, which are deposited interstitially in the rocks, suggests transportation by an unusual solvent, which has very little effect, except addition, on the rocks within the ore itself. Because the added elements resemble the trace element suite in petroleum, it has been postulated by some as the ore fluid (Erickson and others, 1953). Liquid CO₂ has been characterized as a non-polar, or weakly polar solvent (Mellor, p. 58) and conceivably might be a solvent for a similar suite of metals, as well as for organic materials. Furthermore, recent laboratory experiments indicate that the solubility of certain crude oils in liquid CO₂ is great enough to warrant its use in secondary oil recovery. (Johnson and others, 1952).
The natural gases of the Colorado Plateau are well known as tending to run very high in CO₂ (table 1). In reading an account of the distribution of CO₂-rich gas wells in the United States, one is struck by the number of occurrences in southwestern Colorado, northwestern New Mexico, and southeastern Utah. By coincidence, perhaps, a dry ice plant was planned in Grand Junction to utilize CO₂-rich gas from the Morrison formation.

In summary, we have felt for some time that there is a "gimmick" in the vexing problem of the origin of the ores of the Colorado Plateau. From the little we know, CO₂ is just sufficiently common and unusual to lead us to the suspicion that it will bear a good hard look as a candidate for ore depositing honors.

REFERENCES


