graphic projection (Fig. 4, right) across several of the bands. By the criteria of Raleigh (7), the slip system was identified as T equal to (100); t equal to [001] (8). This is the most common glide system in the sheared olivine.

Preferred orientation apparently has been induced in those pellets sheared at high normal pressures. The x-ray powder transmission pattern, taken normal to the pellet surface, shows the characteristic arcuate pattern caused by preferred orientation. No evidence of recrystallization has been found, probably because the temperature was too low and the heating time too short.

The experiments demonstrate that a significant weakening mechanism, operating at temperatures higher than 300°C, causes a marked change in aggregate mechanical properties when the dunite is associated with even very small amounts of a hydrous mineral.

In addition to the reduction of normal pressure by serpentine dehydration, Raleigh and Paterson (9) suggest that the presence of new phases and new pores in grain boundaries may lead to a decrease of "cohesive strength" in the solid framework of the rock.

The classical Coulomb fracture theory for the generation of earthquakes requires prohibitively high shear stresses to overcome frictional resistance and shear strength. However, it is possible

Oxygen Isotope Fractionation in the

Clayton and Epstein (1) and Engel

et al. (2) analyzed five coexisting pairs

of quartz and dolomite from the Lead-

ville formation and showed that the

oxygen isotopic compositions of each

mineral of a pair are essentially the

same (within 3.5 per mil). These au-

thors suggested that if the isotopic

properties of quartz and dolomite were

indeed similar, dolomite-calcite pairs

could provide a useful geothermometer

with about the same sensitivity as the

quartz-calcite pair. This argument pre-

System Dolomite-Calcite-Carbon Dioxide

that at depths of 20 to 40 km the shear strength of rocks may be greatly reduced by dehydration effects.

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Abstract. Oxygen isotopes were equilibrated between carbon dioxide and

calcite at four temperatures in the range 350° to $610^{\circ}C$ and between carbon

dioxide and dolomite at 350° and 400°C. Carbon of unusual isotopic composition

was used as a tracer to demonstrate the nature and extent of the exchange

process. Extrapolation of these data at lower temperatures indicates that at

25°C dolomite is enriched in oxygen-18 by 6.8 per mil with respect to calcite.

This result indicates that those natural dolomite-calcite assemblages which show

very small fractionations were not formed in isotopic equilibrium.

there is a basic incompatibility in the natural data and at least one of the types of rocks was not formed in isotopic equilibrium.

Whether or not the large fractionations found in the high-temperature assemblages are equilibrium fractionations can be resolved only by equilibration studies in the laboratory. Unfortunately, dolomite is very resistant to isotopic exchange with aqueous solutions in reasonable times. The first attempts to determine the equilibrium fractionations between dolomite and calcite were made by Epstein, Graf, and Degens (4). These authors synthesized dolomite-calcite mixtures by reacting calcium carbonate with magnesium chloride solutions and by decomposing magnesian calcite in the presence of water. The isotopic compositions of the starting materials were not such that values of the final fractionation factor could be bracketed (that is, equilibrium would be approached from only one direction). In addition, there may be unknown kinetic fractionation effects associated with the chemical reactions involved. The fractionations observed in the temperature range 400° to 600°C were approximately 2 per mil, in agreement with the natural hydrothermal results.

Northrop (5) and Northrop and Clayton (6) have derived a dolomitewater fractionation curve by graphically interpolating, to 100-percent exchange, data from experiments in which equilibration was incomplete. The temperature range studied was 300° to 510°C. Although different dolomites gave consistently different results, the combination of the average values with those of the previously determined calcitewater system (7) yielded a dolomitecalcite fractionation curve which extrapolates to a value of 5.6 per mil at 25°C. Due to the uncertainty involved in the extrapolation of the high-temperature data at 25°C, a range of 4 to 7 per mil was given for the dolomitecalcite fractionation at this temperature. These authors also studied carbonate-carbonate and carbon dioxidecarbonate systems but were unable to obtain consistent fractionations.

Our primary aim in this study was to determine dolomite-calcite equilibrium fractionations at some experimentally convenient temperatures in order to resolve the natural data incompatibilities previously mentioned. The only unambiguous way to determine an equilibrium fractionation factor be-

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dicts a dolomite-calcite fractionation of 6 to 10 per mil at 25°C and 1 to 2 per mil at 500°C.

A succession of studies of natural dolomite-calcite assemblages ensued, with conflicting interpretations (3). Essentially, the problem involves the existence of large dolomite-calcite fractionations (2 to 4 per mil) in hightemperature rocks and small fractionations (0 to 1 per mil) in low-temperature rocks. Since isotopic fractionations increase with decreasing temperature,

tween two substances is to carry out true exchange reactions. These are reactions in which only isotopes are exchanged and no phases are either produced or destroyed in the process. In such reactions the attainment of equilibrium can be demonstrated by choosing starting materials with isotopic differences on either side of the equilibrium value. Our approach was to equilibriate dry carbon dioxide gas with each carbonate and derive the dolomitecalcite fractionation by the relation:

1000 ln $\alpha_{dol-eal} =$

1000 ln $\alpha_{\rm CO_2-ca1}$ – 1000 ln $\alpha_{\rm CO_2-do1}$

where α is the fractionation factor, defined below.

The equilibration vessels were made of nickel and each was equipped with a glass stopcock and standard taper joint for ready manipulation on a vacuum line. Temperature was monitored by a Chromel-Alumel thermocouple welded to the bottom of the vessel. The carbonates used were natural hydrothermal samples and the dolomite was stoichiometric and well ordered. The charge consisted of 4 to 5 g of very fine, freshly ground carbonate and 50 to 100 μ mole of CO₂ (dried by passing over a dry-ice trap). Care was taken to ensure that all the solid carbonate rested at the bottom of the vessel. Before the CO_2 was added, the vessel containing the carbonate was heated to 300°C and evacuated to remove traces of moisture. At temperatures of 400°C and below, the reactions were quenched in seconds by pulling the vessel from its furnace and plunging it into cold water. At higher temperatures, the vessels were subjected to a blast of compressed air before being immersed in water, and quench times were of the order of a minute. The CO2 was extracted immediately and its volume was checked manometrically before isotopic analysis.

On the average the oxygen content of the CO_2 amounted to only 0.2 percent of the oxygen in the system. However, during the times of these experiments the CO₂ equilibrated with only a part of the solid carbonate. Haul and Stein (8) have shown that for these exchange reactions there is an initial rapid exchange with the surface of the carbonate, followed by a slow diffusioncontrolled exchange process. We have observed the same rate behavior in this study. Therefore, it was our intention to make the carbonate surface-oxygen to CO₂-oxygen ratio very large to maximize the CO₂ exchange rate, and then 8 APRIL 1966

to demonstrate that the observed fractionation effects were representative of the crystalline carbonate.

Most simply, if this observed rateleveling with time is indeed diffusioncontrolled, there can be no "surface problem," since the process of diffusion necessarily leads to the true equilibrium fractionation, and the same fractionation factor is attained by approach from opposite sides of the equilibrium value. In addition, a simple mass balance calculation shows that penetration several molecular layers into the crystal must have taken place to account for the amount of exchange occurring in some of the experiments in which the starting isotopic compositions of either carbon or oxygen of the CO₂ were vastly different from the carbonate. To illustrate this effect, a sample of CO_2 with a carbon isotopic composition 25 per mil lighter than that of a dolomite was equilibrated with it at 400°C for over 300 hours. After equilibration the carbon isotopic composition of the CO₂ was almost identical with that of the dolomite. Aside from the fact that there would not be enough surface carbon to accommodate this large change, this means that with only surface exchange the dolomite surface should be left isotopically light. A new sample of CO_2 with the same isotopic

Table 1. Oxygen and carbon isotope fractionations between CO₂ and calcite.

Experiment	T(°C)	Δt (hours)	1000 ln $\alpha(O^{18})$	1000 ln α (C ¹³)
29	610	0 95	+2.4 +6.3	-24.7 +1.0
30	610	0 148	+10.0 +6.5	0.0 +3.4
15	520	0 60	$^{+10.0}_{+7.0}$	0.0 + 1.5
16	520	0 220	-1.2 + 6.7	-22.1 -10.9
40	520	0 203	+6.7 +7.2	41.8 2.4
27	400	0 385	+10.0 + 8.6	0.0 +1.5
31	400	0 215	+6.6 +7.9	-10.7 -3.2
41	400	0 325	+13.7 + 8.9	-41.9 -3.2
47	400	0 189	+8.9 +8.2	-3.2 + 3.3
66	350	0 200	+9.1 + 8.9	-3.2 + 1.8

Table 2. Oxygen and carbon isotope fractionations between CO_2 and dolomite.

Experiment	T(°C)	Δt (hours)	1000 ln $\alpha(O^{18})$	$1000 \ln \alpha (C^{13})$
49	400	0 76	+4.3 +6.7	+6.0 +5.6
51	400	0 46 602	+4.2 +5.3 +6.2	+6.0 +5.8 +5.2
59	400	0 245	+10.6 +6.6	-0.1 + 3.9
60	400	0 333	+7.3 +6.3	-25.1 + 0.4
65	400	0 812 2920	+8.5 +7.2 +6.4	-7.6 + 0.5 + 0.6
61	350	0 508 1284	+10.7 +9.7 +8.6	0.0 + 3.0 + 3.1
62	350	0 508	+7.7 +7.1	-22.0 -3.3
63	350	0 812 2920	+9.2 +8.1 +7.8	-5.0 +3.1 +3.1
64	350	0 812 2920	-1.3 + 3.5 + 5.1	-22.0 -2.6 -1.3

Table 3. Equilibrium values of 1000 ln α (O¹⁸). Cal., calcite; Dol., dolomite. Values in parentheses were obtained by extrapolation.

<i>T</i> (°C)	CO ₂ -Cal.	CO ₂ -Dol.	DolCal.
610	6.4	(5.3)	(1.1)
520	7.0	(5.7)	(1.3)
400	8.2	6.5	1.7
350	8.9	7.0	1.9
25	11.4	(4.6)	(6.8)

composition as the bulk dolomite was then equilibrated with this dolomite sample at 350°C for 1300 hours, and the carbon isotopic composition became 0.5 per mil heavier (the oxygen changed 2 per mil to an equilibrium value in the process). Obviously the "exchangeable surface" was not isotopically light (otherwise the CO₂ would have become lighter instead of heavier), which demonstrates that considerably more than the crystal surfaces are involved in these exchange reactions.

The experimental fractionations are listed in Tables 1 and 2 and the inferred equilibrium fractionations at various temperatures are listed in Table 3. The fractionations are presented in terms of 1000 ln α , where α is the O¹⁸/O¹⁶ or C^{13}/C^{12} ratio in CO_2 divided by the same ratio in the carbonate. All isotope ratios were determined mass spectrometrically on carbon dioxide gas. The phosphoric acid fractionation factors used in the analyses of the carbonates are 1.01008 for calcite and 1.01090 for dolomite (9).



Fig. 1. Oxygen-18 fractionation curves in the dolomite-calcite-CO₂ system.

The initial isotopic differences between CO_2 and the carbonate are given at $\Delta t = 0$. The final and in most cases the equilibrium differences (fractionations) are then presented with the length of the run. For each temperature, isotopic equilibrium is demonstrated by approaching it from opposite directions. In those cases where three times are listed, the first analysis of the gas indicated incomplete equilibration and the gas was returned to the reaction vessel for a longer run.

The optimum conditions for the oxygen isotope exchange experiments were: (i) large surface carbonate to CO_2 ratio, (ii) initial oxygen isotopic differences very close to equilibrium, and (iii) initial carbon isotopic differences very far from equilibrium (10). Experiment 40 illustrates this approach. Although the oxygen isotopic composition of the CO₂ changed only 0.5 per mil, the carbon isotopic composition changed over 39 per mil, thus indicating that dynamic exchange was taking place. Experiments 63 and 64 were identical in all respects other than initial isotopic differences. Complete exchange was not attained even after 4 months, owing primarily to the initial distance from equilibrium (as in experiment 61). However, by extrapolating these data to 100-percent exchange by the method of Northrop and Clayton, an equilibrium value of 7.0 is inferred, in excellent agreement with 7.1, the result of the equilibrium experiment 62.

Values of 1000 ln α are plotted against T^{-2} in Fig. 1. The use of T^{-2} rather than T^{-1} has been justified by the theoretical considerations of Urey (11) and Bigeleisen and Mayer (12) and has been used in the presentation of such data for other systems (7, 13). Unfortunately dolomite begins to decompose around 450°C at the pressures of CO_2 used in these experiments (~ 0.3 atm), and at temperatures below 350°C the rates of exchange between CO₂ and carbonates are prohibitively small. Therefore, the dolomite temperature study was restricted to only two points. Two additional points on the CO_2 -calcite curve are known: 0 at T = ∞ and 11.4 at $T = 25^{\circ}$ C. The CO₂calcite fractionation at 25°C is known experimentally from the determinations of the calcite-water (14) and CO_{2} water fractionations (15). Therefore, the CO₂-calcite curve must exhibit considerable curvature with a maximum. However, over the limited temperature range of this study the CO₂-calcite data

fit a straight line very well, and the two CO₂-dolomite points define another straight line. From these lines is derived an expression for the dolomitecalcite fractionation:

1000 ln $\alpha_{dol-cal} = 0.56 (10^6 T^{-2}) + 0.45$

The CO₂-carbonate curves cannot be extrapolated to low temperatures because of their curvature. However, it is the CO_2 that is mainly responsible for this curvature (see CO₂-O data calculated by Urey in 1947, 11) and it is highly probable that two such similar species as these carbonates would have similar isotopic properties and exhibit a linear dolomite-calcite fractionation curve. It is on this basis that the extrapolated value of 6.8 per mil for the dolomite-calcite fractionation at 25°C is considered meaningful. It is questionable what error should be applied to the low-temperature value obtained from the extrapolation of the hightemperature results. But even if it were as much as 2 per mil, the conclusions would not be affected.

Because the dolomite data were restricted to such a small temperature range, the dolomite-calcite expression above is not considered accurate enough for use in oxygen isotope thermometry. Nonetheless, when applied to the data of Clayton and Epstein (1), reasonable temperatures of formation are obtained, showing that the original postulation of these authors is essentially correct.

The results of this study in conjunction with those of Northrop and Clayton (6) demonstrate that the large number of low-temperature dolomitecalcite assemblages exhibiting small oxygen isotope fractionations are not equilibrium assemblages.

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Rotation of the Planet Mercury

Abstract. The equations of motion for the rotation of Mercury are solved for the general case by an asymptotic expansion. The findings of Liu and O'Keefe, obtained by numerical integration of a special case, that it is possible for Mercury's rotation to be locked into a 2:3 resonance with its revolution, are confirmed in detail. The general solution has further applications.

The recent discovery by Pettengill and Dyce (1) that Mercury's rotation is direct, with a period approximately twothirds of its period of revolution about the Sun, has led to speculation (see Colombo, 2) that there might be a resonance lock. Liu and O'Keefe (3)independently investigated this possibility by numerical integration of the equations of motion and found such a resonance lock to be possible.

If A < B < C are the principle moments of inertia of Mercury, assumed constant, and if C is taken perpendicular to the orbital plane, then the equations of motion governing the rotation of Mercury are, from equation 4 of Liu and O'Keefe,

$$\frac{d^2\psi}{dt^2} + \frac{3}{2}\lambda \frac{\sin 2\phi}{r^3} = 0 \qquad (1)$$

where ϕ is the angular displacement of the principle axis, A, from the direction of the radius vector; $\psi = f + \phi$, f being the true anomaly of Mercury; $\lambda =$ (B - A)/C and r is the distance from Mercury to the Sun; and t is time. Units of mass, length, and time are chosen so that Mercury's mean motion and semimajor axis, and the constant of gravity, are all unity.

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Equation 1 can be derived from a Hamiltonian system of two degrees of freedom in the variables ψ , Ψ (conjugate to ψ), ℓ (the mean anomaly of Mercury) and L (conjugate to ℓ), with the equations of motion

$$\frac{d\Psi}{dt} = F^*_{\psi}, \frac{d\psi}{dt} = -F^*_{\Psi}$$

$$\frac{dL}{dt} = F^*_{\ell}, \frac{d\ell}{dt} = -F^*_{L}$$
(2)

the subscript denoting partial differentiation, and

$$F^* = -L - \frac{1}{2} \Psi^2 + \frac{3}{4} \lambda \frac{\cos 2(f - \psi)}{r^3}$$
 (3)

where f and r are considered to be functions of ℓ .

A resonance condition, if it exists, will be due to the extremely slow motion of the angle

$$\psi - \frac{3}{2} \ell$$

and to the small divisors associated with it. Hence it makes sense to isolate this variable by a canonical transformation:

$$x_1 = 2L + 3\Psi, y_1 = \ell/2$$

$$x_2 = \Psi - \frac{3}{2}, y_2 = \psi - \frac{3}{2}\ell$$

with equations of motion

$$\frac{dx_i}{dt} = F_{y_i}, \frac{dy_i}{dt} = -F_{x_i}, i = 1,2$$
 (4)

and Hamiltonian (dropping terms which do not contribute to Eqs. 4)

$$F = -\frac{1}{2}x_1 - \frac{1}{2}x_{2^2} + \frac{3}{4}\lambda \frac{\cos(2f - 3\ell - 2y_2)}{r^3}$$

where f, ℓ , and r are to be considered as functions of y_1 .

To treat this equation we make use of von Zeipel's method of averaging, a method closely related to other averaging techniques but especially suited to canonical systems, as described by Brouwer and Clemence (4). Moser has given rigorous justification of the use of such methods (5).

We introduce a new set of canonical variables, denoted by bars over the corresponding old variables, by means of a determining function

$$S = \Sigma \,\overline{x}_i y_i + S_1 + S_{3/2} + \dots$$

where $S_n = O(\lambda^n)$, λ being considered a small parameter, and

$$x_i = S_{y_i}, \, \overline{y_i} = S_{\overline{x_i}}, \, i = 1,2$$
 (6)

Similarly, the new Hamiltonian, $\overline{F}(\overline{x}_i,$ \bar{y}_i), is developed in a series

$$F = F_0 + F_1 + F_{3/2} + \dots$$

with

 $\overline{F}_n = O(\lambda^n).$

It is assumed that the new variable $\bar{x}_2 = O(\lambda^{\frac{1}{2}})$. (Incidentally, the choice ascending powers of $\lambda^{\frac{1}{2}}$ is not mandatory but is convenient for present purposes.) Then Eqs. 6 are inserted into the equation expressing the constancy of the Hamiltonian under this transformation.

$$F(x_i, y_i) = \overline{F}(\overline{x}_i, \overline{y}_i)$$

which is expanded in powers of $\lambda^{\frac{1}{2}}$, and terms of like order are compared. The result is a sequence of equations for determining F_n and S_n recursively. In this particular case it is possible to choose \overline{F} so that it does not depend on y_1 (making \overline{x}_1 a formal integral of the system) and S so that it is periodic in y_i . The Hamiltonian \overline{F} is also a formal integral. and, according to Moser's work, it is an approximate integral for all time.

Carrying out this process, since $\overline{F}_{3/2}$ happens to vanish, we obtain

$$\overline{F} = -\frac{1}{2} \overline{x}_1 - \frac{1}{2} \overline{x}_2^2 + \frac{3}{4} \lambda \cos 2\overline{y}_2 \times \left\{ \frac{1}{2\pi} \int_0^{2\pi} \frac{\cos (2f - 3l)}{r^3} \, dy_1 \right\} + O(\lambda^2)$$

The integral can be evaluated straightforwardly by the formulas of elliptic motion and is a function of the eccentricity e alone. It is not necessary for e to be a small parameter. For Mercury e = 0.2056, and the quantity in brackets is 0.613, so that the new Hamiltonian is

$$\overline{F} = -\frac{1}{2} \bar{x}_1 - \frac{1}{2} \bar{x}_{2^2} + 0.460\lambda \cos 2\bar{y}_2 + O(\lambda^2) \quad (7)$$

Excluding the constant first term and the terms in $O(\lambda^2)$, we recognize that this expression is like the Hamiltonian of a pendulum, with stable equilibria at $y_2 = 0, \pi$ and unstable equilibria at $y_2 = \pm \pi/2$. The limiting period P of the oscillation around the stable equilibria is found to be

$$P_{\rm lim} = 0.178 \ \lambda^{-1/2}$$
 years. (8)

By standard pendulum formulas, if the maximum value of y_2 in its oscillation about $\overline{y}_2 = 0$ is ϕ , the period is given by a factor containing an elliptic integral:

$$P = P_{\lim} \left\{ \frac{2}{\pi} \int_0^{\pi/2} \frac{d\theta}{(1 - \sin^2 \phi \, \sin^2 \theta)^{1/2}} \right\} \quad (9)$$

For the value $\lambda = 5 \times 10^{-5}$ investigated by Liu and O'Keefe, P_{1im} turns out to be 25.1 years, agreeing with their result of 25 years. They found that the amplitude of oscillation of the instantaneous period was 0.008 days, which

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