

Reports

Phase Changes in the Upper Mantle

Abstract. *The C-region of the upper mantle has two transition regions 75 to 90 kilometers thick. In western North America these start at depths of 365 kilometers and 620 kilometers and involve velocity increases of about 9 to 10 percent. The locations of these transition regions, their general shape, and their thicknesses are consistent with, first, the transformation of magnesium-rich olivine to a spinel structure and, then, a further collapse of a material having approximately the properties of the component oxides. The velocity increases associated with each transition region are slightly less than predicted for the appropriate phase change. This can be interpreted in terms of an increasing fayalite content with depth. The location of the transition regions and the seismic velocities in their vicinity supply new information regarding the composition and temperature of the upper mantle. The depths of the transition regions are consistent with temperatures near 1500°C at 365 kilometers and 1900°C at 620 kilometers.*

At a depth near 400 km in the earth's mantle there is a sudden increase in the gradients of the seismic velocities. There is also evidence that the increases with depth of density, electrical conductivity, seismic quality factor, and viscosity are most rapid somewhere near this depth. The gradients of seismic velocities are abnormally high between about 350 and 700 km of depth when compared with the effects expected from simple compression. It has been proposed that solid-solid phase changes are responsible for the anomalous properties of this section of the mantle. In particular the olivine-spinel phase change has received much attention in discussion of this region. The great breadth of this zone, the rate of velocity increase, the large total change in velocity, the details of the velocity variation with depth, and zero pressure density of the lower mantle, however, indicate that other factors must also be operating.

In the Jeffreys-Bullen model of the earth the transition region is spread over the interval 413 to 983 km and the velocity and density gradients, although high, decrease with depth in this region. Plausible phase changes in materials thought to be important for the mantle would lead to narrower transition regions and higher velocity gradients than observed. Furthermore, the rate of increase of velocity would be uniform or increase slightly with depth, at least in the olivine-spinel reaction (1).

The zero-pressure density of spinel is about 10 percent greater than the density of olivine, and the difference decreases with pressure. The total density increase across the transition zone of the mantle is about 20 percent. Birch and Ringwood (2) have suggested that other phase changes may be involved, both to spread out the transition and to increase the ultimate density. In particular Birch (2) speculated that the lower mantle consisted of dense, closely packed polymorphs of normal ferromagnesian silicates having closely packed oxide structures. Although spinel is about 10 percent denser than olivine, the silicon atoms in spinel are tetrahedrally coordinated, so it is far from closely packed. It can therefore be expected that spinel will transform at sufficiently high pressure to denser phases where the silicon is octahedrally coordinated. This would involve a further density increase of about 8 percent.

Surface wave studies (3) have shown that the abnormally high velocity gradients in the transition region are concentrated in two relatively narrow zones 50 to 100 km thick, instead of being spread out uniformly over some 600 km, as was previously thought. This fact has important implications regarding the composition and physical state of the upper mantle. Several recent studies using travel times and apparent velocities of body waves have verified the presence of these two transition regions (4). The most detailed of these

are the studies of Johnson, and of Archambeau and Flinn. These two structures are shown in Fig. 1. CIT 204 is from Johnson and is appropriate for the western part of North America. CIT 109P is from Archambeau and Flinn and is appropriate for the western part of the midcontinent region of the northern United States for depths between 400 and 800 km. CIT 11CS3, also shown in Fig. 1, is an earlier model from Niazi and Anderson which is based on both surface waves and apparent velocities of compressional waves (4).

For the present discussion it is the similarities between the models rather than the differences which are important. In all cases there is a very rapid increase in velocity between about 100 and 150 or 175 km and major discontinuities starting at 320 to 365 km and 620 to 640 km. The new mantle model of Johnson, CIT 204, is based on the most complete set of data and will form the basis of most of the following discussion. The regions of rapid velocity increase are between 365 and 440 km, and 620 to 700 km. The velocity gradients in the adjacent sections of mantle are appropriate for normal compression. This includes the section of mantle between 440 and 620 km which is in the middle of what has been designated as the transition region. The boundaries between regions B, C, and D of the upper mantle can now be placed near 365 km and 700 km, at least for western North America. This agrees with surface wave results (5) which represent world-wide average conditions. Bullen, using earlier velocity models, placed the boundaries of the transition region at 413 and 984 km. Regions B and C also have to be further subdivided. Extending Bullen's nomenclature we have B', subcrustal layer; B'', low-velocity zone; B''', region of normal velocity gradients; C', transition layer (365 to 440 km); C'', region of normal velocity gradients (440 to 620 km); and C''', transition layer (620 to 700 km). Present evidence suggests that the location and details of the subdivisions of region B vary laterally.

The transition regions are 75 to 90 km thick. The total increase in velocity associated with each of the two transitions is about 9 to 10 percent. If there is no change in composition in this region these velocity jumps would each correspond to about 7 percent increase in density. The total density jump in this region which is required to satisfy

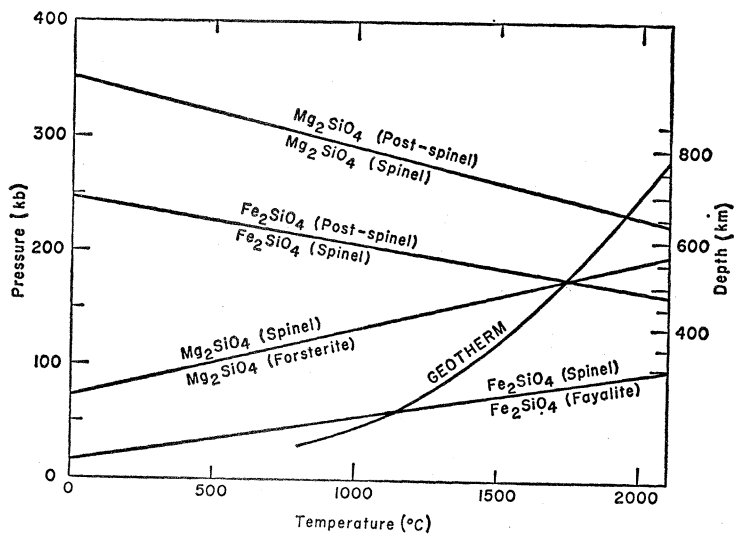
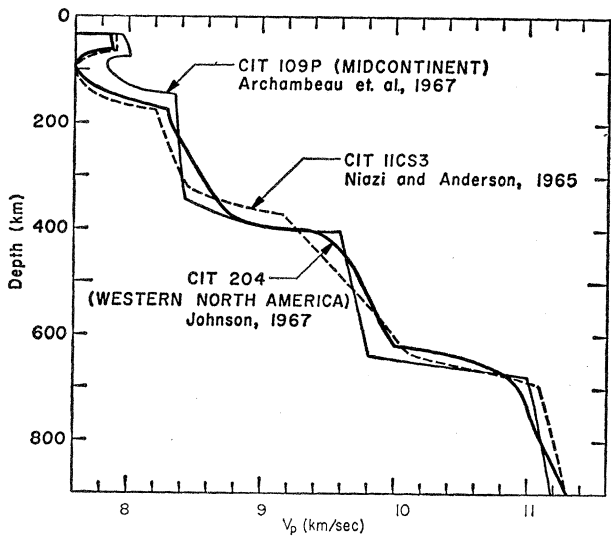


Fig. 1 (left). Compressional velocity versus depth in the upper mantle.

Fig. 2 (right). Equilibrium lines for olivine-spinel and spinel-oxides transformations in Mg_2SiO_4 and Fe_2SiO_4 .

the earth's moment of inertia and the periods of free oscillation, however, is slightly greater than 20 percent. The density increases associated with the olivine-spinel transformation and the spinel-oxides decomposition at transition depth pressures are about 8 percent and 7 percent, respectively. The implication is that the major portion of the density increase in the transition

zone can be attributed to solid-solid phase changes.

Akimoto and Fujisawa (6) studied olivine-spinel solid solution equilibria in the system $Mg_2SiO_4-Fe_2SiO_4$ to pressures of 92 kb and compositions to 60 mole-percent Mg_2SiO_4 . Simple extrapolation permitted them to estimate the olivine-spinel transitions pressure in pure Mg_2SiO_4 at 150 ± 10 kb at $800^\circ C$.

This is in good agreement with other estimates (7, 8). The new NaCl pressure scale of Jeffrey *et al.* (9) will reduce this pressure estimate by 20 to 30 percent.

Furthermore their work indicated that the width of the two-phase region in olivine with 70 to 90 percent Mg_2SiO_4 and 30 to 10 percent Fe_2SiO_4 would be about 20 to 30 kb which in the upper

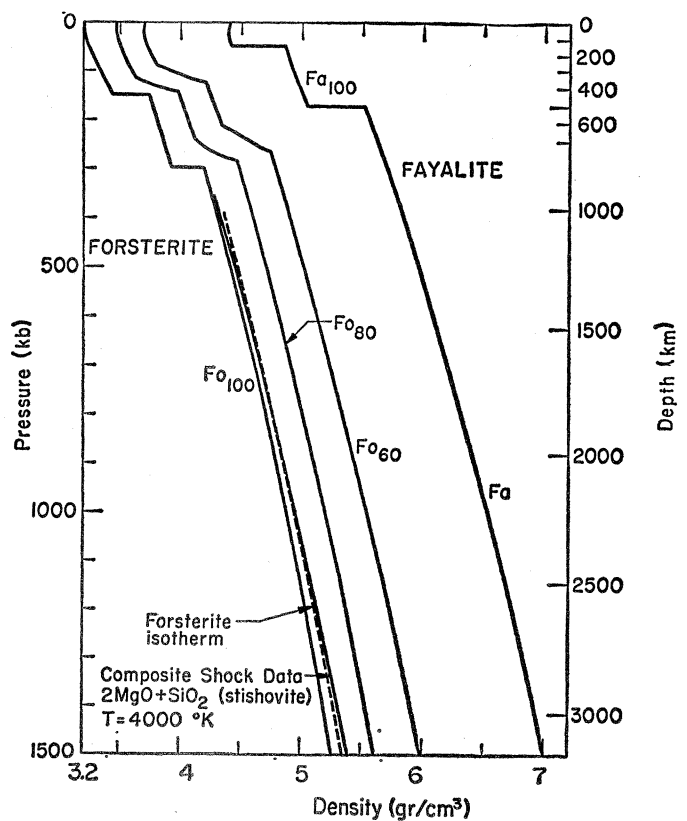
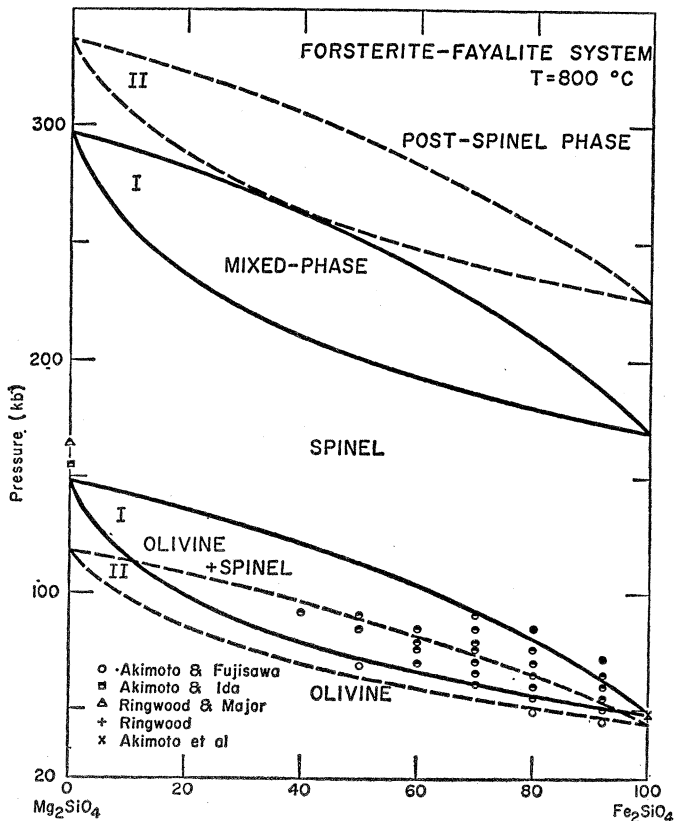


Fig. 3 (left). Estimated phase diagram for system $Mg_2SiO_4-Fe_2SiO_4$ using values for parameters from Tables 1 and 2. Calculations I and II are based on different pressure scales.

Fig. 4 (right). Density as a function of pressure and depth in the earth taking into account temperature, pressure, and solid-solid phase changes for olivine of four different compositions (22). Locations and details of transition regions computed at $800^\circ C$.

mantle would correspond to a depth interval of 60 to 90 km. The good correspondence between the pressure and width of the transition found in the laboratory and the seismic transition region near 400 km encouraged us to make a more detailed comparison. The experimental parameters of the olivine-spinel phase change supply the heat of formation of spinel which is required in order to estimate the transition parameters of the spinel-oxides transition. We will assume that the spinel will ultimately break down to a phase composed of MgO, FeO, and SiO₂ with the silicon in octahedral coordination. We assume that this phase has approximately the same thermochemical properties as the oxides.

Akimoto *et al.* (10) determined the equation of the transition curve for the fayalite-spinel transition as $P = 12 + 0.045T$, where P is in kilobars and T is in degrees centigrade. These experiments were performed in the pressure range 47 to 67 kb. According to the new NaCl pressure scale of Jeffrey *et al.* (9), pressure estimates in this range should be revised downward by about 10 percent. This revision gives $P = 15 + 0.036T$ as the equation of the fayalite-spinel transition. Similarly, the estimate of 155 kb for the transition of forsterite to spinel at 800°C (8) must be revised downward by 20 to 30 percent.

Table 1 summarizes the thermochemical data for the reactants in the system MgO • SiO₂ • FeO. The transition pressures and slopes of the reaction lines were computed from this data by requiring that the Gibbs free energy change be zero at the reaction and by the use of the Clausius-Clapeyron equation. Results are given in Table 2 and Fig. 2. The change of volume and slope of the reaction line are tabulated both at zero pressure and at the transition pressure. The uncertainties in the calculated pressures are at least 10 percent.

We treat the mantle as an ideal binary system Mg₂SiO₄-Fe₂SiO₄. If we assume constant temperature the boundaries of the two-phase region depend only on P_1 and P_2 and $2RT/\Delta V_1$ and $2RT/\Delta V_2$ where P_1 and P_2 are the transformation pressures of pure Mg₂SiO₄ and Fe₂SiO₄ and ΔV_1 and ΔV_2 are the corresponding changes in molar volumes going from the olivine to the spinel phase (1).

Similarly the phase diagram for the spinel-postspinel transition can be calculated from the parameters in Table

Table 1. Thermochemical data for silicates and oxides. The data are based primarily on compilation by Clark (20).

Silicates and oxides	M (g/mole)	ρ (g/cm ³)	V (cm ³ /mole)	ΔH°_{298} (kcal/mole)	S°_{298} (cal/mole °K)
Mg ₂ SiO ₄ forsterite	140.7	3.214	43.79	-520.3	22.73
Mg ₂ SiO ₄ spinel	140.7	3.536	39.79*	-512.8†	17.70‡
MgO periclase	40.3	3.584	11.25	-515.0§ -143.7	6.44
SiO ₂ stishovite	60.09	4.287	14.02	-205.9	6.13**
2 MgO + SiO ₂	140.7	3.855	36.50	-493.3	19.01
Fe ₂ SiO ₄ fayalite	203.79	4.405	46.26††	-353.5	34.7
Fe ₂ SiO ₄ spinel	203.79	4.849	42.03††	-353.0‡‡	31.3‡
FeO wüstite	71.9	5.969	12.05	-353.2§§ -63.8	13.74
2 FeO + SiO ₂	203.79	5.346	38.12	-333.5	33.61

* Based on ΔV (forsterite → spinel) = -4.0 cm³/mole, Akimoto and Fujisawa (6).
 † Based on olivine-spinel transition parameters $P_{298} = 110$ kb, $\Delta V^P = 2.8$ cm³ where superscript P refers to transitional pressure.
 ‡ From entropy-density correlation for spinels.
 § Based on olivine-spinel transition parameters $P_{298} = 74$ kb, $\Delta V^P = 3.1$ cm³. ** Ahrens and Syono (21).
 †† Akimoto *et al.* (10). ‡‡ Based on olivine-spinel transition parameters $P_{298} = 12$ kb, $\Delta V^P = 4.0$ cm³. §§ Based on olivine-spinel transition parameters $P_{298} = 16$ kb, $\Delta V^P = 4.0$ cm³.

2. The postspinel phase is taken to be a homogeneous ideal solid-solution. Stishov (11) speculates that the crystal structure of this phase may be an imperfect structure of the NaCl type. The high-pressure experiments of Albers and Rooymans (12) on the spinel FeCr₂S₄ yielded an apparently homogeneous phase related in structure to smythite, a defect NaCl structure. We assume that the postspinel phase is a homogeneous phase having the thermochemical properties of the component oxides, periclase, wüstite, and stishovite.

Calculations were performed for two sets of parameters; calculation I is based on the original data, primarily of Akimoto and Fujisawa (6). Calculation II represents an attempt to take into account the new NaCl pressure scale of Jeffrey *et al.* (9). The phase diagrams are given in Fig. 3. Also shown are the original data of Akimoto and Fujisawa (6) on the system Mg₂SiO₄-Fe₂SiO₄ and Akimoto and Ida (8), Ringwood (7), and Ringwood and Major (7) on the pure end members, extrapolated in some cases to 800°C.

In calculation I the mixed phase region thickness is 37 to 42 kb or 105 to 120 km for the olivine-spinel reaction for the compositional range 80 to 60 mole-percent Mg₂SiO₄. In calculation II the mixed phase region is 23 to 26 kb or 65 to 74 km thick for the same range in composition. The effect of a temperature gradient would be to increase

the thickness of the transition region. The thickness of the transition region will be decreased if the iron content at the base of this zone is greater than at the top (Fig. 3). This would also serve to increase the density through this region and to decrease the overall velocity increase.

The mixed phase region between the spinel and a postspinel phase having the properties of the component oxides is spread over a depth interval of about 110 to 135 km (calculation I) or 85 to 105 km (calculation II) over the same composition range. The negative slope of the transition line combined with a geothermal gradient of 1°C per kilometer will reduce the thickness of this transition region by about 17 km. The temperatures in the mantle are, of course, unknown but estimates range from about 1400° to 1800°C at 400 km and 1600° to 2000°C at 600 km. The pressure of the olivine-spinel transition then would be about 143 ± 20 kb (I) or 130 ± 20 kb (II) for 80 Mg₂SiO₄ • 20 Fe₂SiO₄ and about 123 ± 20 (I) or 114 ± 20 (II) kb for 60 Mg₂SiO₄ • 40 Fe₂SiO₄. The uncertainties include both the probable range of temperatures and uncertainties in the slope of the reaction line but not the uncertainty in the 800°C pressure estimates. These pressures correspond to depths of about 426 ± 60 km (I) and 388 ± 60 km (II) for 80 percent forsterite and to depths of 368 ± 60 km (I) and 342 ± 60 (II) for

Table 2. Transition parameters at 298°C. Superscripts 0 and P refer to zero pressure and transition pressure, respectively. Values in parentheses are estimates based on NaCl pressure scale of Jeffrey *et al.* (9).

Transitions	P (kb)	($\Delta P/\Delta T$) ⁰	($\Delta P/\Delta T$) ^P
Mg ₂ SiO ₄ (olivine) → Mg ₂ SiO ₄ (spinel) ΔV ⁰ = 4.00 ΔV ^P = 2.8 (3.2)	110 (74)	+0.053	+0.06
Mg ₂ SiO ₄ (spinel) → Mg ₂ SiO ₄ (post-spinel) ΔV ⁰ = 3.29 ΔV ^P = 2.2 (2.2)	310 (350)	-.017	-.06
Fe ₂ SiO ₄ (olivine) → Fe ₂ SiO ₄ (spinel) ΔV ⁰ = 4.20 ΔV ^P = 4.0 (4.0)	13 (16)	+ .034	+ .04
Fe ₂ SiO ₄ (spinel) → Fe ₂ SiO ₄ (post-spinel) ΔV ⁰ = 3.91 ΔV ^P = 3.0 (3.0)	190 (245)	-.025	-.04

60 percent forsterite, in good agreement with the depth to the top discontinuity. The predicted depths to the top of the lower discontinuity are 546 ± 60 km (I) and 670 ± 60 km (II) for 80 percent forsterite and 473 ± 60 km (I) and 620 ± 60 km (II) for 60 mole-percent forsterite. These depths are again in good agreement with the location of the lower discontinuity. Considering the uncertainties in the calculations, the depths and thickness of the predicted transition regions are in very good agreement with the seismic discontinuities for compositions and temperatures thought to be appropriate for the mantle.

From the phase diagram, Fig. 3, and the known or estimated densities of the various phases as a function of pressure and composition, it is possible to compute the variation of density throughout the mantle, including the two mixed phase regions. We use a semi-empirical equation of state (13) and assume hydrostaticity to compute density as a func-

tion of composition, temperature, and pressure. The parameter of this equation of state is the isothermal pressure derivative of the bulk modulus and is taken to be 4.096 in the upper mantle and transition region and 3.0 in the lower mantle. The results are shown in Fig. 4 for forsterite, fayalite, and two intermediate compositions which bracket the presumed composition of the mantle. The temperatures used are similar to the lower estimates of Clark and Ringwood (14). Representative values are 704°C at 100 km, 1390°C at 300 km, 1567°C at 400 km, and 1841°C at 600 km. The temperature gradient in the upper mantle is high enough to generate a region of decreasing density down to about 30 km. The depths to the transition regions are for 800°C. According to Table 4 the upper discontinuity will be deeper and the lower discontinuity will be shallower at mantle temperatures. The agreement between the theoretical depths, thicknesses, and

shapes of the transition regions and the new seismic results is remarkable.

Since it is the compressional velocity, V_P , rather than the density which is measured directly, we use Birch's (15) empirical relationship

$$V_P = a(\bar{M}) + 3.31 \rho$$

which connects compressional velocity to density and mean atomic weight. The velocities so computed are given in Fig. 5 for two different compositions and a range of temperatures at the transition depths.

The compressional velocities for the mantle model of Johnson are also shown. Note the remarkable similarity between the theoretical and experimental velocities, both in the self-compressed and transition regions. The absolute values of the velocities and the depths to the transition regions are consistent with a mantle composition between about 60 and 80 mole-percent forsterite; the fayalite content appears to increase with depth from 500 to 700 km.

An increase of fayalite with depth is not inconsistent with electrical conductivity measurements. The electrical conductivity in the upper mantle may increase by about 3 orders of magnitude near 400 km (16), although other interpretations are possible. The electrical conductivity jump associated with olivine-spinel phase change in fayalite is about 2 orders of magnitude (see 17). Another order of magnitude increase can be obtained by increasing the fayalite content from 20 to 30 mole-percent (18).

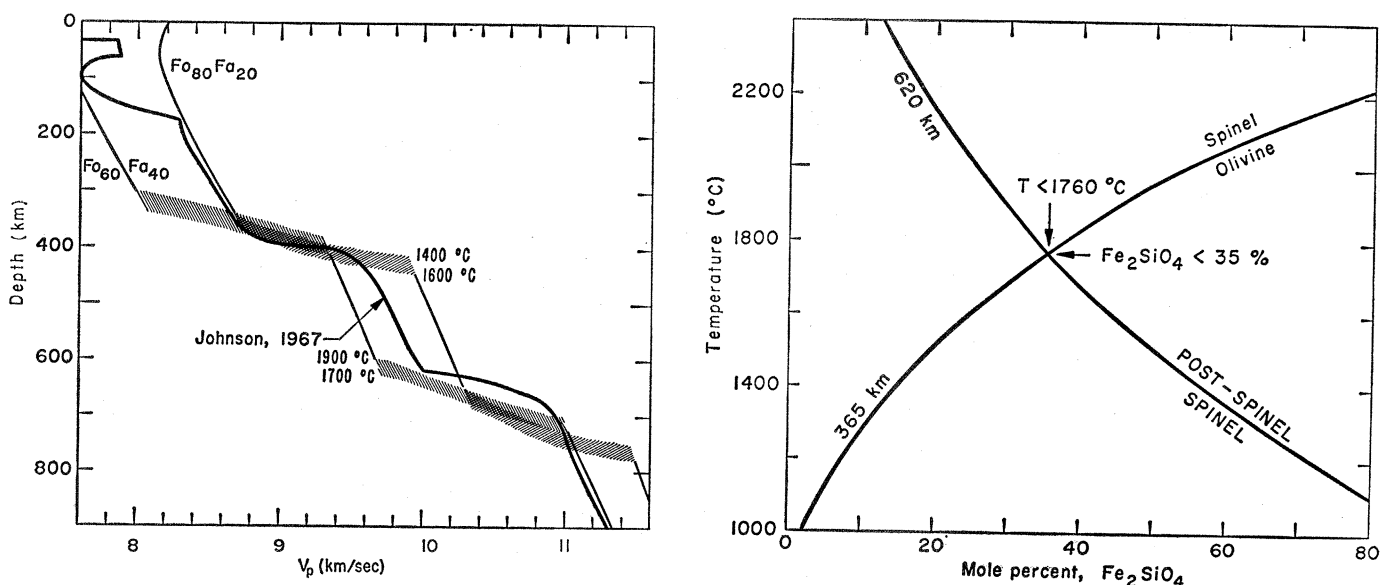


Fig. 5 (left). Compressional velocity, V_P , as a function of depth for $Fo_{80}Fa_{20}$ and $Fo_{60}Fa_{40}$ compared with seismic velocity structure of Johnson (4). Fig. 6 (right). Temperature and compositions in the mantle that are consistent with locations of seismic discontinuities at depths of 365 and 620 km.

Minor components such as Al_2O_3 and CaO may be important, but their effect on the location of the phase boundaries and the seismic velocities is difficult to evaluate. A garnet content of 10 percent will decrease the velocity in the spinel portion of the mantle by 0.1 to 0.3 km/sec and by 0.2 to 0.6 in the postspinel portion of the mantle. Everything else being equal, the presence of 10 percent garnet will decrease the velocity below 700 km by about 0.2 to 0.3 km/sec more than the velocity is decreased between the two discontinuities.

The effect of garnet, or of an assemblage containing Al_2O_3 in addition to SiO_2 , MgO , and FeO , on the transition pressures is not known. However, the good correspondence between the depths and thicknesses of the transition layers in the mantle with those predicted for an olivine mantle suggests that the major features of the mantle between 200 and 700 km are dominated by phase changes in the system Mg_2SiO_4 - Fe_2SiO_4 .

The depths to the top of the transition regions are governed both by composition and temperature. Using the phase diagram, Fig. 3, and the values of the dP/dT from Table 2 it is possible to construct curves that give the temperatures and compositions that are consistent with the observed depths of the seismic discontinuities; Fig. 6 shows the results. Taking the depths to the tops of the discontinuities as 365 and 620 km and dP/dT of the transitions as $+0.05$ kb/ $^\circ\text{C}$ and -0.05 kb/ $^\circ\text{C}$, respectively, upper bounds can be put on both the fayalite content and the temperature at the top of the upper discontinuity by assuming that the temperature increases with depth. From Fig. 6 Fe_2SiO_4 is less than 35 percent and T is less than 1760°C . For a fayalite content in the transition region of 20 mole-percent and uniform, $T(365)$ equals 1520°C and $T(620)$ equals 2160°C . For a fayalite content at 620 km of 30 mole-percent, $T(620)$ equals 1880°C .

The depth distribution of earthquakes has been studied by many seismologists (19) but there has been no obvious correlation with known discontinuities or low-velocity zones. The maximum depth of earthquake activity is near 700 km. Gutenberg computed the average annual seismic energy release as a function of depth and found maxima centered at 350 and 600 km. There is thus some suggestion that the depth distribu-

tion of deep earthquakes in tectonic regions is influenced, if not controlled, by the location of transition regions. One possibility is that stress is most effectively concentrated in those regions where the elastic properties are changing most rapidly. Another possibility is that phase changes are still occurring in the mantle in tectonic regions and the stress field in the mantle is readjusting to these phase changes.

Recent high pressure experimental petrology studies on the olivinespinel transition in the $\text{Mg}_2\text{SiO}_4 \cdot \text{Fe}_2\text{SiO}_4$ system have made it possible to construct a theoretical mantle that is in remarkable accord with latest seismic data. Although the mantle certainly contains minerals other than olivine, the important features of the velocity variation in the transition region seem to be controlled by solid-solid phase changes in magnesium-rich olivine. There is some evidence that the fayalite content increases with depth in the transition region, although other interpretations are possible. The depths of the transition regions are consistent with temperatures near 1500°C at 365 km and 1900°C at 620 km.

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Abundance of Type II Diamonds

Abstract. More than 1300 natural microdiamonds, mostly averaging 0.0015 carat, were studied for transparency to ultraviolet radiation, $\lambda 2537$. Of most of them, of this size, 16 percent were completely transparent (presumably type II); 27 percent, completely absorbent (presumably type I). The remainder transmitted partially. The number of type-II diamonds is unexpectedly very high; a considerable proportion of microdiamonds may begin life as type II, later incorporating absorbent type-I material.

Diamond is such a variable material that, for conclusions about general properties, large numbers must be examined. Examination of tens (1) or even of 100 or 200 (2) may at times be inadequate. In a recent investigation (3) of birefringence, 5000 separate diamond crystals were examined. Considerations of time and manpower obviously restrict the kinds of studies when the numbers of crystals involved run into thousands. We now report an ultraviolet-absorption investigation made on 1310 selected, optically very clear, visually perfectly transparent, natural microdiamonds.

Synthetic industrial diamonds vary from 0.2 to 1.0 mm in diameter. As some of these are under optical examination, we have set out to study the