Effects of Iron/Magnesium Ratio on *P*- and *S*-Wave Velocities in Olivine

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Velocities of both the compressional and the shear waves measured on seven synthetic olivines with different Fe/Mg ratios are reported. The olivine compositions studied were 100% Fo, 95% Fo, 90% Fo, 85% Fo, 80% Fo, 50% Fo (where Fo stands for forsterite), and 100% fayalite. Effects of the Fe/Mg ratio on the compressional, shear, and bulk velocities, the bulk modulus, and the seismic parameter are examined for olivine at ambient conditions. An iron substitution in the olivine lattice results in a systematic decrease in these velocities, the seismic parameter, and the bulk modulus such that, for fayalite as compared with forsterite, there is a 22% decrease in V_p, a 30% decrease in V_s, a 16% decrease in V_Φ, a 30% decrease in seismic Φ, and only about a 4.7% decrease in K_s. The iron substitution increased Poisson's ratio of olivine by 19% for fayalite relative to forsterite. It was also found that the bulk modulus of olivine decreases almost linearly with increases in the iron content in the olivine lattice.

Experimental information on the elastic-wave velocities in olivines of the forsterite-fayalite series is essential to a better understanding of the physics and chemistry of the upper mantle. Literature dealing with elastic properties of olivine is abundant. *Adams* [1931] studied effects of iron content on compressibility of olivine. *Verma* [1960] reported the complete set of single-crystalline elastic constants determined on gem-quality periodot crystals. *Birch* [1960, 1961*a*], *Simmons* [1964], *Kanamori and Mizutani* [1965], and *Christensen* [1966] measured velocities in various dunites under pressures up to 10 kb. *Birch* [1962] reported S velocity in dunite as a function of temperature to about 800°K at 9 kb. *Schreiber and O. L. Anderson* [1967] studied the behavior of P and S velocities to about 2 kb on a synthetic forsterite sample with 6% porosity; on a similar specimen, *Soga and O. L. Anderson* [1967] reported velocities as a function of temperature to about 1000°K. *Trunin et al.* [1965] and *McQueen et al.* [1967] published shock-compression data on various dunites. More recently, *Graham and Barsch* [1969] and *Kumazawa and O. L. Anderson* [1969] reported both pressure and temperature derivatives at room temperature of the elastic constants for single-crystal forsterite. While the present work was in progress, data on the compressional velocity in fayalite have been reported by *Mizutani et al.* [1970]. *Fujisawa* [1970] also reported both the compressional- and the shear-velocities measured in a polycrystalline fayalite sample with 2.5% porosity. However, except for the work of *Adams* [1931], none of these investigators studied effects of Fe/Mg ratio on the elastic properties of olivine. In the literature, many authors (see, for example, *Birch* [1961*a*, *b*], *McQueen et al.* [1964], *Press* [1968*b*], and *Christensen* [1968]) called attention to the need of a study for effects of iron on the elasticity of olivine.

In his measurements on the effect of pressure on the volume of 'pure fayalite,' Adams found 1.04 as the bulk modulus of fayalite having a density of 4.068 gm/cm³. On the assumption that Poisson's ratio is 0.27, Adams then found 6.6 km/sec as the velocity of P waves for fayalite at ambient conditions. The crystal density of fayalite is 4.393 g/cm³ [*Yoder and Sahama*, 1957; *Robie et al.*, 1966] and suggests Adams's fayalite sample may not have been a representative sample of fayalite. The fayalite sample used by Adams was recently discussed by *Birch* [1969, p. 33]; Birch suggested a redetermination of elastic properties on a fayalite sample with the correct density.

In this paper, we report new data on the compressional (P) and shear (S) wave velocities

determined on artificially produced olivines with widely varying Fe/Mg ratio. Effects of the Fe/Mg ratio on velocities, bulk modulus, and seismic parameter are studied for these olivines at ambient conditions. The results reported here should be of interest to the scientific community concerned with the physical state and chemical composition of the earth's upper mantle, since they represent the only such systematic data existing for the forsterite-fayalite series. Sample preparation and experimental procedures are described in section 1. In section 2, experimental data are presented for 7 olivines with different Fe/Mg ratios. Although our velocity data reported here are at ambient conditions only, their relative variations in velocities as the Fe/Mg ratio changes may be useful in correlating to the first order with the velocities of elastic waves within the earth's mantle, determined from seismological data. On this basis, a brief discussion related to the earth is made in section 3.

1. Samples and Experimental Procedure

1.1 Preparation and Characterization of Specimens

Forsterite. Starting with stoichiometric amounts of MgO and SiO_2 powders, forsterite was prepared by solid state reaction at 1350°C for 120 hours. The starting powders were obtained as follows: SiO_2 (cristobalite) was prepared from reagent grade silicic acid (Baker Chemical Company) by heating for 12 hours at 1300°C. Microscopic examinations revealed that over 80% of the particles had mean diameter of about 5–15μ, about 10% of the particles were less than 5 μ, and about 10% were greater than 15 μ size. A similar fraction of MgO was obtained by milling AR-grade periclase powders (Mallinckrodt Chemical Works). The stoichiometric amounts of these powders were hand mixed in acetone, and the process was continued as the acetone evaporated. The powders were then dried and heated in a temperature-controlled, platinum-wound furnace for periods up to 120 hours at 1350°C. The extent of solid-state reaction was determined by use of x-ray diffraction and microscopic observations. The forsterite powders thus prepared were then crushed and milled to about 1–10 μ in particle size; they were then hot-pressed into a dense-formed disk with a procedure described earlier [see *Crandall et al.*, 1961].

Fayalite. Starting with stoichiometric amounts of SiO_2 and iron oxalate, fayalite was prepared by solid-state reaction under a controlled atmosphere. The reaction temperature was 1200°C for 72 hours. SiO_2 (cristobalite) as a starting material was prepared as above (see 'Forsterite') from reagent grade silicic acid by heating. The ion oxalate (Amend Drug and Chemical Company, Inc.) was used as the source of 'FeO' because iron in iron oxalate is in the ferrous state. In addition, under solid-state reaction with silica, the iron oxalate generates a slightly reducing atmosphere, e.g.,

$$SiO_2 + 2FeC_2O_4 \cdot 2H_2O + O_2 \rightarrow Fe_2SiO_4 + 4CO_2 + 4H_2O$$

This reducing condition is essential in the formation of fayalite. X-ray diffraction and microscopic examination were used as before to characterize the formed fayalite. The fayalite powders thus prepared were then crushed and milled down to about 1–10 μ in particle size; they were hot-pressed into a dense-formed disk.

Forsterite-fayalite solid solutions. Our synthesis of the olivine solid solutions followed a procedure used by *Yoder and Sahama* [1957] and also by *Akimoto and Fujisawa* [1968]. In brief, starting with ready-made powders of forsterite and fayalite crystallites, the desired proportions of the two end-member olivines were made to react at 1200°C under a controlled atmosphere for 12 hours. Five solid-solution olivines thus prepared are 95% Fo, 90% Fo, 85% Fo, 80% Fo, and 50% Fo where '% Fo' designates a mole per cent forsterite. X-ray diffraction and microscopic examinations were used throughout to characterize each reaction product. All the reaction products used in the present work were found to be mostly (better than 95% by volume) a single-phase olivine of the specified composition. The resistant hot-pressing method was used for making dense-formed disks of these olivine specimens.

The hot-pressed disks were then appropriately cut,[1] ground, and polished to give rectangular

[1] Most hot-pressed samples show apparent anisotropies in their elastic properties, an effect often due to a preferred orientation of mineral grains and to the orientation of pores (including

TABLE 1. Composition and Measured Properties of Various Olivine Samples (at 296°K and 1 bar)

Sample Composition, mole %	Sample Density, g/cm³	Porosity, %	v_p, km/sec	V_s, km/sec	k_1	k_2
100 Fo	3.164	1.65	8.459 (±0.5)*	4.938 (±0.4)*	0.9655	1.1923
95 Fo/5 Fa	3.176	2.98	8.287 (±0.6)	4.823 (±0.4)	0.9619	1.2148
90 Fo/10Fa	3.270	1.90	8.226 (±0.4)	4.769 (±0.4)	0.9583	1.2381
85 Fo/15Fa	3.386	2.83	8.088 (±0.6)	4.676 (±0.4)	0.9556	1.2560
80 Fo/20Fa	3.365	2.19	8.017 (±0.5)	4.615 (±0.4)	0.9529	1.2744
50 Fo/50Fa	3.732	1.80	7.454 (±0.5)	4.178 (±0.4)	0.9362	1.3947
100 Fa	4.287	2.41	6.502 (±0.5)	3.421 (±0.4)	0.9011	1.7031

* Uncertainties in per cent.

prisms having two opposing faces that are parallel to each other. The density of each specimen was determined by measuring weight in air and in Nujol, a mineral oil (Plough, Inc.), whose specific gravity at 22°C is 0.8815 g/cm³. The sizes of all specimens are in the neighborhood of 1 cm³, large enough to make acoustic measurements on them in all the three mutually perpendicular directions.

Table 1 lists the composition and density of each specimen. The porosity in the specimen was calculated from the X-ray density [see *Yoder et al.*, 1957; *Akimoto et al.*, 1968] of the corresponding chemical composition of olivine.

1.2. Experimental Method

Experimental method used here was identical to our earlier work on quartz and rutile [*Chung and Simmons*, 1969]. Reviewing in brief, the measurements of sound velocities were made with the pulse-echo-overlap method. X-cut and AC-cut quartz transducers with resonance frequencies of 20 MHz were used for generation of *P* and *S* waves, respectively. The material used for acoustic bonding between specimen and transducer was a 50% (by volume) mixture of phthalic anhydride and glycerine. The pressure system used here was a simple piston-cylinder setup of standard design [see *Brace et al.*, 1969]; the sample was not jacketed. The pressure medium was reagent-grade petroleum ether. The pressure was read directly from a precalibrated 7500-bar Heise gauge. The readability of this gauge is better than 0.2%.

2. Experimental Results and Discussion

2.1. Data

The primary data determined in our ultrasonic experiments were the pulse-repetition frequencies $F_i(p)$ in each specimen for both *P* and *S* waves at 296 (±1)°K as a function of hydrostatic pressure to about 7.5 kb. From these $F_i(p)$ data, sound velocities at zero-pressure were found by extrapolation of high-pressure data back to the zero-pressure point (see, for example, *Mizutani et al.* [1970], p. 2743). These values represent crack-free but not pore-free velocities. Values of *P* and *S* velocities evaluated at zero-pressure are entered in the 4th and 5th columns of Table 1, and an estimated total uncertainty in the velocities is indicated. This uncertainty includes estimated experimental errors and variations in velocities due to an apparent anisotropy of the specimen. The apparent anisotropy of each specimen was observed to be small, possibly because of the way our specimens were prepared (see footnote 1). The anisotropy was less than 0.3% of the velocities listed in Table 1.

As is seen from Table 1, our samples contain about 2 to 3% porosity. Isotropic elastic properties at zero-porosity must be evaluated from acoustic measurements made on our porous samples. *Weil* [1964, p. 217] and *Walsh* [1965] discussed how the elastic properties of nonporous polycrystalline aggregates can be evaluated from elastic data obtained on a porous aggregate. We used, as in our earlier work with rutile and quartz [*Chung and Simmons*, 1969, p. 135], the Weil-Hashin relation for the shear and bulk

cracks). The anisotropy of a polycrystalline specimen produced by hot-pressing methods can be reduced by cutting a sample out of the hot-pressed disk at the 45° angle from the direction of hot-pressing and by subsequent heat treatment of the sample.

moduli with constants k_1 and k_2. Numerical values of k_1 (for the shear modulus) and k_2 (for the adiabatic bulk modulus) found for our samples are tabulated in the 6th and 7th columns of Table 1, and they were used to obtain the zero-porosity elastic properties of various olivines. The zero-porosity elastic properties of olivine as a function of the Fe/Mg ratio are summarized in Table 2.

2.2. *Comparison with Literature Data*

The elastic parameters of various olivines widely used in the geophysical discussion are tabulated in Table 3. These include experimental values measured on natural olivine rocks and synthetic aggregate-samples as well as gem- and they are probably not representative of Tables 2 and 3 reveals the following:

1. *For forsterite,* the present elasticity data are consistent (within the stated experimental uncertainties) with the corresponding data cited under references *d* and *e*, Table 3. In light of these data, the elastic properties of forsterite reported in references *a*, *b*, and *c* are too low and they are probably not representative of the intrinsic properties of forsterite.

2. *For peridot* (with about 93% Fo), literature data cited in references *f* and *g* are slightly higher than the present values. It is interesting to note the bulk modulus of peridot (with about 93% Fo) according to references *f* and *g* is slightly higher than that of forsterite. The result of the present work indicates that, contrary to the observation of *f* and *g*, the value of the bulk modulus decreases slightly with increasing the Fe/Mg ratio in olivine.

3. *For hortonolite* (with about 50% Fo), the present data somewhat differ from values reported in references *l* and *m*. The apparent difference may be associated with the composition of the samples studied by the authors cited as they are reflected in the sample densities. Additional data for a hortonolite sample from the type locality in Monroe, New York, became available in the literature since the present paper was submitted for publication. *Mao et al.* [1970] reported *P* and *S* velocities of hortonolite dunite ($\rho = 3.934$ g/cm^3) with the ($Fo_{37}Fa_{57}Te_6$) composition as 7.46 and 4.05 km/sec, respectively. These new data appear to be quite consistent with the present data for 50% Fo after an allowance for the slightly higher iron content. As discussed by Mao et al., the lower values of *P* and *S* velocities for the Mooihoek dunite measured by *Birch* [1960] and *Simmons* [1964] may be due to alteration to serpentine.

4. *For fayalite,* the present data differ by a few per cent from the literature values cited in references *n*, *o*, and *p*. For example, values of the bulk modulus as reported by *Adams* [1931], *Mizutani et al.* [1970], and *Fujisawa* [1970] are 1.04, 1.10, and 1.164 mb, respectively, and these values may be compared with 1.22 mb found from the present work.

3. Generalization and Discussion

Some generalization about the elastic properties in the forsterite-fayalite solid-solution series can be made from Table 2. An iron substitution in the olivine lattice results in a systematic decrease in the velocity of both *P* and *S* waves. The iron increases the density but reduces the bulk modulus of olivine (thus

TABLE 2. Zero-Pressure Elastic Properties of Olivine as a Function of Fe/Mg Ratio (at 296°K)

Property	Unit	Olivine Composition, mole %						
		100% Fo	95% Fo	90% Fo	85% Fo	80% Fo	50% Fo	100% Fa
ρ_0	g/cm^3	3.217	3.273	3.330	3.386	3.440	3.780	4.393
$\bar{m}$	gram	20.12	20.58	21.00	21.48	21.93	24.60	29.10
V_p	km/sec	8.534	8.422	8.317	8.216	8.116	7.534	6.637
V_s	km/sec	4.977	4.892	4.815	4.739	4.663	4.213	3.494
μ	mb	0.797	0.783	0.772	0.760	0.748	0.671	0.536
K_s	mb	1.281	1.277	1.274	1.272	1.269	1.251	1.220
σ_s		0.242	0.245	0.248	0.251	0.254	0.273	0.308
Φ	(km/sec)^2	39.8	39.0	38.3	37.6	36.9	33.1	27.8
V_Φ	km/sec	6.309	6.245	6.189	6.132	6.075	5.753	5.273

TABLE 3. Literature Data on the Elastic Properties of Various Olivines (evaluated at 296°K and 1 bar)

Property	Forsterite				Peridot			Dunite (at 10 kb)			Hortonolite			Fayalite		
ρ, g/cm^3	3.222[a]	3.021[b]	3.204[c]	3.224[d]	3.222[e]	3.324[f]	3.311[g]	3.312[h]	3.326[i]	3.330[j]	3.300[k]	3.595[l]	3.744[m]	4.068[n]	4.393[o]	4.283[p]
V_P, km/sec	(8.1)	7.586	8.16	8.569	8.593	8.482	8.421	8.42	(8.42)	8.66	8.52	7.4	7.36	6.60	6.74	6.485
V_S, km/sec	···	4.359	4.90	5.015	5.032	4.933	4.887	(4.83)	4.83	4.74	4.80	···	3.90	···	···	3.417
Φ, (km/sec)^2	···	32.2	35.0	39.9	40.1	38.8	39.1	39.8	39.8	44.8	41.9	···	33.9	···	···	···
K_S, mb	1.22	0.974	1.12	1.286	1.291	1.289	1.294	1.32	1.32	1.50	1.38	1.12	1.27	1.04	1.10	1.164
μ, mb	···	0.574	0.769	0.8108	0.816	0.806	0.7908	···	0.776	0.75	0.760	···	0.57	···	···	0.513
σ_S	(0.27)	0.254	···	0.240	···	0.245	0.246	0.255	0.255	0.286	0.267	(0.27)	0.30	(0.27)	(0.28)	···

Values in parentheses are assumed values by the author cited.

[a] *Adams* [1931]; natural forsterite minerals.

[b] *Schreiber and Anderson* [1967]; sintered sample.

[c] *Marsh and Sheinberg* [1969]; hot-pressed sample.

[d] *Kumazawa and Anderson* [1969]; VRH values (arithmetic average of Voigt and Reuss values) from single-crystal data.

[e] *Graham and Barsch* [1960]: VRH values from single-crystal data.

[f] *Verma* [1960]; VRH values from single-crystal data of $(Mg_{0.92}Fe_{0.08})_2SiO_4$.

[g] *Kumazawa and Anderson* [1969]; VRH values from single crystal data of $(Mg_{0.93}Fe_{0.07})_2SiO_4$.

[h] *Birch* [1960]; Twin Sisters dunite (92% olivine + 7% pyroxene).

[i] *Simmons* [1964]; Same as *Birch's* [1960] samples.

[j] *Christensen* [1966]; Twin Sisters dunite.

[k] *Kanamori and Mizutani* [1965]; Horoman dunite (88% olivine + 11% pyroxene).

[l] *Adams* [1931]; 50% Fo + 50% Fa.

[m] *Birch* [1960] and *Simmons* [1964]; Mooihoek Mine hortonolite dunite.

[n] *Adams* [1931]; sample obtained from a blast-furnace slag.

[o] *Mizutani et al.*, [1970]; polycrystalline sample.

[p] *Fujisawa* [1970]; polycrystalline sample.

making Fe-rich olivine more compressible) because the bulk modulus is inversely proportional to volume [see, for example, *O. L. Anderson* and *Nafe*, 1965; *Knopoff*, 1967; *D. L. Anderson and O. L. Anderson*, 1970]. The velocity varies with the inverse square root of the density but it varies as the square root of the modulus-to-density ratio. In the forsterite-fayalite solid-solution series, an increase in the iron content affects only very slightly the bulk modulus (i.e., 1.281 mb for forsterite and 1.22 mb for fayalite) but causes a wide variation in the density.

The compressional velocity in olivine as a function of density is plotted in Figure 1. The contours in the figure are derived from Birch's law [*Birch*, 1961*b*, 1964; *Simmons*, 1964] relating the velocity-density-mean atomic weight of rocks and minerals. Note that the compressional velocity in olivines with different Fe/Mg ratios lie on a dashed line of the forsterite-fayalite isomorph. Not only the compressional velocity as shown in Figure 1, but also the shear velocity and the bulk velocity (though not shown here) were seen to behave in a similar manner for this series of olivine isomorphs.

Olivines are unstable at high pressures and transform to spinel structures with an increase of density to about 10% [see *Ringwood*, 1969]. At 1273°K, the olivine-spinel transformation pressures for forsterite and fayalite are 140 kb and 50 kb, respectively [see *Ringwood* and *Major*, 1966; *Ringwood*, 1969; *Akimoto and Fujisawa*, 1968]. The determination of the sound velocities in the olivine-transformed spinels with the compositions studied here has been an important objective of our laboratory efforts. Thus far, however, we were unable to obtain enough amounts of the transformed spinel materials. While the present report was in preparation, an important paper by *Mizutani et al.*, [1970] was published. These authors, after their successful preparation of a fayalite-transformed spinel sample, performed a measurement of the compressional velocity as a function of pressure to about 6 kb. They found the compressional velocity in the spinel form of Fe_2SiO_4 is 8.05 km/sec at ambient conditions. This value is an approximate 20% increase in the velocity from that of fayalite.

The important datum of *Mizutani et al.* [1970] has been plotted in Figure 1. This datum

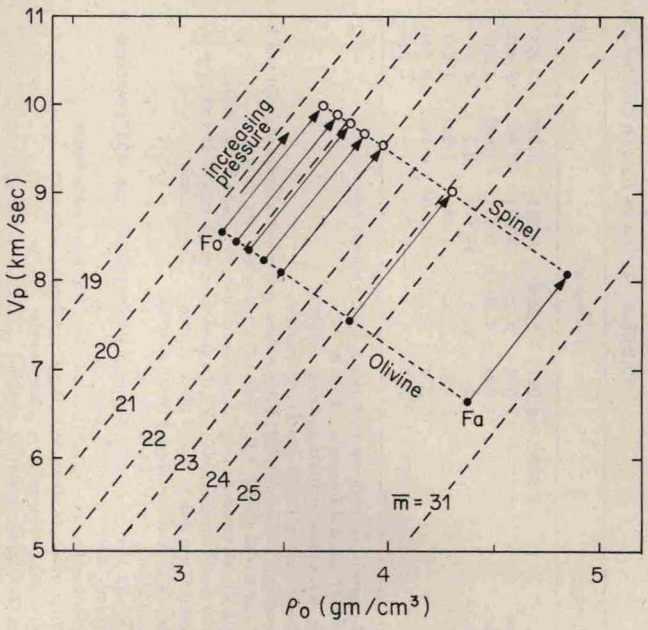

Fig. 1. The compressional velocity–density relation for olivine with different Fe/Mg ratios. The contours are drawn from Birch's law. The closed circles represent experimental quantities; the open circles show hypothetical data points.

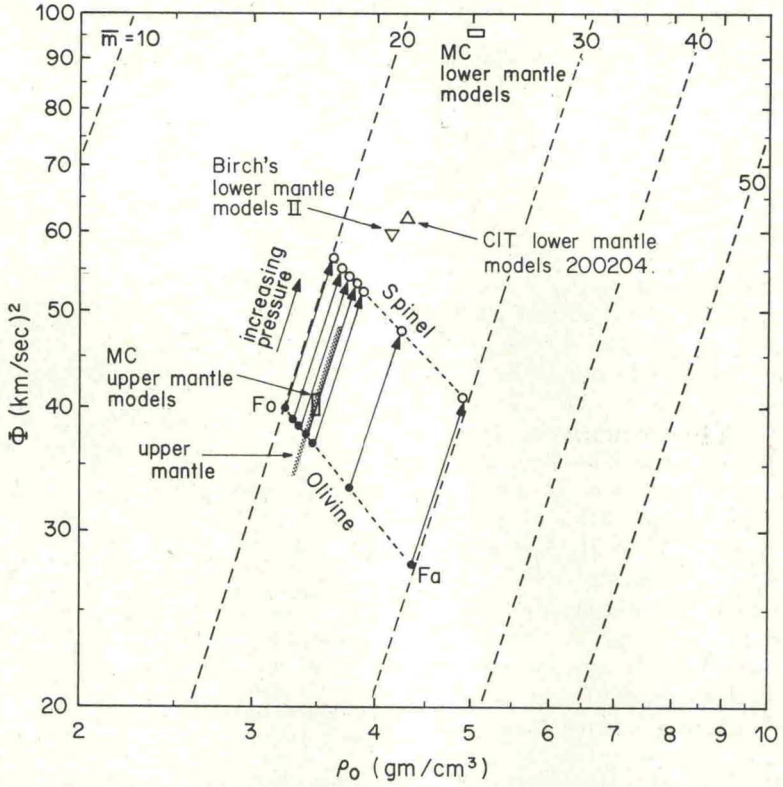

Fig. 2. The seismic parameter–density relation for olivine with different Fe/Mg ratios. The contours are derived from D. Anderson's Φ law. The closed circles show experimental quantities; the open circles show hypothetical data points. Various models of the upper and lower mantles are indicated in the figure. The range of Monte Carlo (MC) successful models is shown by rectangles.

point meets at the intercept of the two lines drawn from the density and the mean atomic weight surprisingly well.[2] If we were to assume that the olivines of other compositions behave similarly, a series of lines parallel to a specific mean atomic weight characterized by the Fe/Mg ratio in the olivine composition can be drawn. Knowing the end-point density of the spinel of the given composition, then one is able to estimate velocity in that spinel phase. In this manner, both the compressional- and the shear-wave velocities in the spinel phase were found; they are plotted in Figure 1 for the compressional-wave velocity as an example. These hypothetical data points are identified in the figure with open circles.

The geophysical significance of these estimated data points, though they are subjected to confirmation, should not be underestimated. As noted by *D. L. Anderson* [1967*a*], recent progress in seismological advances (surface waves, free oscillations, large seismic arrays studies, etc.) make it possible to refine the standard velocity-depth profiles of Bullen, Gutenberg, Jeffreys, and Lehmann. One of the more significant contributions is a refinement of the velocity-depth profiles in Bullen's *C*- region ranging from about 400 km down to 1000 km in depth. Surface wave studies by *D. L. Anderson and Toksöz* [1963] demonstrated, for example, that this region consists of a series of relatively thin

[2] Both D. L. Anderson and O. L. Anderson, during their respective presentations at the Birch symposium in April 1970, made similar observations. On the basis of data presented by *Mizutani et al.* [1970], the same observation was also noted by *Liebermann* [1970] in a paper that has just appeared in this journal. The writer expresses his thanks to R. C. Liebermann for sending a copy of his paper along with a review as a referee for the present report.

(about 50 km thick) regions of a rapid increase in velocity. *D. L. Anderson* [1967*a*] further noted that the most notable of these discontinuities occur at depths near 400 and 600 km. Most of the discontinuities can be attributed to solid-solid phase changes that have been directly observed in the laboratory or inferred from studies of the behavior of analog compounds or shock-wave studies [see, for a recent review, *Ahrens et al.*, 1969]. From the laboratory measurements, it has been inferred that near 400 km olivine collapses to the spinel or 'pseudo-spinel' phase with 8% (for Fe_2SiO_4) to 10% (for Mg_2SiO_4) increase in density.

The seismic parameter–density–mean atomic weight relation for olivine is shown in Figure 2. The contours of the mean atomic weights drawn in the figure are derived from Anderson's Φ law [*D. L. Anderson*, 1967*b*]. (See also *D. L. Anderson* [1969] for a modification). Figure 2 shows the effect of the Mg-Fe substitution in the olivine solid-solution system. Clearly shown here is that the iron substitution in the olivine lattice is very sensitive to the seismic parameter. The seismic parameters inferred from the estimated velocities of spinels are entered also in Figure 2 and identified with open circles. The widely inferred models of the upper and lower mantles [*Birch*, 1961*b*, 1964; *Clark and Ringwood*, 1964; *Anderson*, 1967*b*; *D. L. Anderson and Smith*, 1968] are indicated in the figure. The range of Monte Carlo successful models for the upper and lower mantles given by *Press* [1968*a*, *b*] is also shown by rectangles in the figure. It is seen that the present data on the seismic parameters obtained for olivine with about 83% Fo agree very well with Press's upper-mantle models.

Although olivine may be the most abundant mineral in the upper mantle, a more realistic discussion of the petrology of the mantle from the elasticity studies must include information on the elastic properties of pyroxene and garnet and their variations with pressure and temperature for these minerals. In a subsequent series of communications, we shall report these data; only then will a discussion of the elasticity and composition of the mantle be attempted.

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