Ramp compression of magnesium oxide to 234 GPa

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Abstract. Single-crystal magnesium oxide (MgO) samples were ramp compressed to above 200 GPa pressure at the Omega laser facility. Multi-stepped MgO targets were prepared using lithography and wet etching techniques. Free surface velocities of ramp-compressed MgO were measured with a VISAR. The sound velocity and stress-density response were determined using an iterative Lagrangian analysis. The measured equation of state is consistent with expectations from previous shock and static data as well as with a recent X-ray diffraction measurement under ramp loading. The peak elastic stresses observed in our samples have amplitudes of 3-5.5 GPa, decreasing with propagation distance.

1. Introduction
Periclase, MgO, is of fundamental importance for geophysics as the end-member of the (Mg,Fe)O solid solution. The Earth’s lower mantle encompasses about 50% of the planet and reaches up to 135 GPa pressure. Experimental studies of expected lithologies show that (Mg,Fe)O along with (Mg,Fe)SiO3 perovskite are the dominant mineral phases of Earth’s lower mantle, with ferropericlase amounting to ~20-25% of the lower mantle by volume [1]. At higher pressures, MgO is also of interest for understanding the cores of giant planets in our solar system [2] as well as possible super-Earth planets orbiting other stars [3,4]. In addition to its geophysical and planetary importance, periclase is of fundamental interest as a prototypical ionic solid with a simple crystal structure and a wide stability field. As a result, MgO has been extensively studied by static compression, shock compression, and theoretical methods. MgO has many attributes of an ideal pressure standard for static high-pressure experiments. A number of recent studies [5–8] have used experimental and theoretical elasticity and equation of state data to construct equations of state of MgO suitable for use as pressure standard in diamond anvil cell experiments. As static experiments more routinely extend into the 100-300 GPa range, MgO will likely remain one of the most important reference materials and better knowledge of its equation of state and properties at ultrahigh pressures is a priority. In this work, we report a study of MgO using laser-driven ramp compression [9–11]. Ramp compression achieves high compression...
at relatively modest temperatures [12]. This technique enables us to explore new regions of thermodynamic space lying between the principal isentrope and Hugoniot. The refractory nature of MgO means that ramp compression experiments will be able to probe MgO in the solid state up to very high compression. Here, we report quasi-isentropic equation of state data of MgO ramp compressed up to 234 GPa.

2. Experiment

Ramp compression experiments were performed using the Omega laser at the Laboratory for Laser Energetics (LLE) of the University of Rochester [13]. In these experiments, a high-powered laser is used to launch a planar compressive wave into the sample. The laser pulse and the corresponding stress history in the sample are approximately ramp shaped with ~5-10 ns duration. Measurement of the free surface velocity (opposite to the loading surface) using a VISAR [14] provides information about the stress response of the target material. By using a stepped sample with variable thicknesses, wave propagation times and hence sound velocities through the sample can be measured. As opposed to shock compression, where a large amount of entropy and heating are produced, ramp compression keeps the temperature relatively low so that the solid state can be maintained up to higher pressures [4,12].

The samples were high purity (>99.95%) single-crystal MgO (3.58 g/cm³) fabricated with a series of ~5-μm thick steps. The targets were prepared using lithography at the Micro/Nano Fabrication Laboratory (MNFL) at Princeton University. We used two-inch diameter single-crystal MgO wafers oriented parallel to [100] (MTI Corp.). Creation of each step in the wafer involved application of a photoresist, followed by removal of material by etching. Etching was performed using a 1:1 ratio of H₃PO₄ and H₂O at 100°C. The process was repeated three times using a mask shifted to different positions to create a stepped sample with four different thicknesses. After etching, the samples were polished to the required thicknesses and cut into individual pieces for experiments. The final dimensions of the samples were 3 x 3 mm square plates with four layers of approximate widths of 0.25-0.3 mm and thicknesses of nominally ~25/30/35/40 μm. The crystals were oriented with the [100] axis along the loading direction. A 0.1-μm W layer was deposited on the stepped surfaces to enhance the reflectivity for wave profile measurements as discussed below. The MgO was then attached to a 20-μm thick and 2 x 2 mm square diamond ablator with a 4-μm thick Au heat shield deposited between the two materials. Sample metrology was performed to characterize surface roughness, thickness gradients, and step heights.

A schematic experimental set-up is shown in figure 1. Each stepped MgO target
assembly was attached to the rear surface of a hohlraum. The hohlraum (General Atomics Corp.) is a gold cylinder with typical dimensions of 2.7-mm diameter, 2.0-mm length, and 25-μm thickness, with a 1.5-mm diameter laser entrance hole. The inner walls of the hohlraum were illuminated with 12-15 beams of the Omega laser with a total energy up to 1-1.6 kJ. The beams were applied in an azimuthally symmetric geometry at incidence angles of 48° and 60°. The laser pulse shape was a composite made up of two individual pulses with a total length of 6.5 ns (figure 2, inset). Lasers heated the walls of the hohlraum and generated a time-dependent and spatially uniform distribution of thermal X-rays within the Au cavity. The characteristic radiation temperature reaches a peak of about 120 eV as measured by an X-ray spectrometer [15]. X-rays impinge on the diamond and ablate its front surface (figure 1). This generates a counter-propagating compression wave moving forward into the diamond and the MgO sample. As the compression wave reaches the back of the sample, the surface accelerates forward resulting in a ramped increase in free surface velocity, \( u_{fs} \). The free-surface velocity as a function of time for each step of the sample was recorded using a line-imaging VISAR (velocity interferometry system for any reflector) [14]. Figure 2 shows a typical example of a measured wave profile (shot #s63791) across four thicknesses of an MgO sample.

3. Analyses and Discussion

Our results can be used to constrain the peak elastic stresses supported by [100] MgO under ramp loading. Figure 3(a) shows a typical wave profile for one step on a MgO sample. The profile shows an initial arrival at \( \sim 0.2-0.3 \) km/s (figure 3a, inset) that is attributed to the elastic precursor of MgO. There is then a rapid rise to a plateau around 3.2-3.5 km/s followed by a second ramped increase to the peak free surface velocity. The second plateau is due to the elastic precursor of the diamond ablator propagating through the MgO sample. The peak elastic stress, \( \sigma_E \) [16], characterizes the maximum amplitude of uniaxial elastic loading the material can support. We observe elastic stress amplitudes of 3-5.5 GPa for our ~25-45 μm thick samples (figure 3b). To calculate these, we assumed that the particle velocity, \( u = u_{fs}/2 \) and the compressional sound velocity in the [100] direction of MgO is 9.1 km/s [17]. The precursor amplitude decreases with increasing sample thickness (figure 3b) and exhibits a weak relaxation behind the initial peak (figure 3a, inset). We can compare these results to the behaviour of MgO under shock loading with mm-thick samples. Gas-gun experiments on [100]-oriented MgO have yielded values of \( \sigma_E \) for MgO of 2.5 GPa (for peak stresses of 5-11 GPa) [17], 3.5-8.9 GPa (for peak stresses of 17-42 GPa) [18] or 2.6-5.3 GPa (for peak stresses of 38-57 GPa) [19] for specimens of 3-8 mm thickness. Polycrystalline MgO samples exhibit lower elastic limit values of 1-1.5 GPa [20]. Our results under ramp loading yield similar peak elastic stresses as those observed on shock loading on single crystals.

Stress and density are calculated from the measured wave profiles using an iterative Lagrangian analysis method [21–26]. The inset of figure 4 shows Lagrangian sound velocities, \( C_L \), as a function of

\[ C_L = \text{constant} \]

**Figure 3.** (a) A typical wave profile for MgO (shot s63791) showing elastic limit signature. The inset shows an enlargement of the profile in of elastic limit region. (b) Elastic limit versus different thickness of the sample from our experiments.
free surface velocity for four experiments and their weighted mean (black curve). The nearly flat response in the \( u_s = 0-3.5 \) km/s range reflects the constant shock velocity as a result of the diamond elastic wave and we treat this as an initial shock \([11]\). Subsequently, the Lagrangian sound speed rises as the ramp wave propagates through the MgO to the peak free surface velocities.

From the Lagrangian sound velocity, stress and density can be obtained from the equations:

\[
\sigma_{xx} = \rho_0 \int_0^u c_L(u) du \tag{1}
\]

\[
\rho = \left( \frac{1}{\rho_0} - \frac{1}{\rho_0} \int_0^u \frac{du}{c_L(u)} \right)^{-1} \tag{2}
\]

where \( \rho_0 \) is initial density of MgO and \( u \) is particle velocity. As a first approximation, it is assumed that particle velocity equals one-half of the measured free surface velocity. This is then corrected iteratively using backward and forward integration in space by a characteristics-based approach enabling the in-material particle velocity, \( u \), to be determined from the measured free surface velocities \([9,25,26]\). The resulting stress-density states are shown in figure 4. The maximum stress reached is 234 GPa.

Diamond anvil cell (DAC) data (red circles) under quasi-hydrostatic conditions up to 118 GPa obtained using a helium pressure media \([30]\) are also plotted in figure 4. Previous shock studies (yellow triangle) \([19,20,27–29]\) are shown as well. The red dotted line extending past the DAC data shows an estimated isotherm up to 250 GPa based on a Vinet equation of state \([31]\) fitted to these static data. The yellow dashed line is the calculated Hugoniot curve from a linear fit to the shock velocity and particle velocity \( (U_s \text{ (km/s)} = 6.68 + 1.35u \) \([19,20,27–29]\)) relationship for MgO. The purple line is the calculated adiabatic compression curve derived from the adiabatic bulk modulus and its isentropic pressure derivative. The initial shock due to the diamond precursor is expected to raise the sample temperature by ~700 K \([32]\) and the final temperature at 234 GPa can be estimated to be a minimum of ~1000 K by assuming isentropic compression above the initial shock. For comparison, the expected temperature along the Hugoniot at 200-230 GPa is ~3700-4600 K. Our ramp-compression data are intermediate between the shock and

![Figure 4. Stress vs. density for ramp compressed MgO. Black curve shows the mean from our four individual experiments. Representative uncertainties for the ramp compression data are one standard deviation. The blue square with error bar shows the result from X-ray diffraction on ramp compressed MgO \([4]\). Shown for comparison are shock data \([19,20,27–29]\) (gold triangles) and room temperature static data \([30]\) (red circles). The red dotted line shows a Vinet equation of state extrapolation of the static data, the yellow dashed line is the calculated Hugoniot curve and the purple line is the calculated principal adiabat. The inset shows calculated Lagrangian sound velocities as a function of free surface velocity for the four experiments and their weighted mean. Longitudinal and bulk sound speeds of MgO are indicated with red and light blue solid circles, respectively.](image-url)
extrapolated static data (figure 4). Although the uncertainties are large, this is consistent with expectations for the likely temperatures achieved in our experiments. Recently it has become possible to carry out direct X-ray diffraction measurements of ramp-compressed samples [33]. In figure 4, the blue square shows the result of an X-ray diffraction measurement on ramp-compressed MgO [4]. Within uncertainty, the results from wave profile analysis and X-ray diffraction are consistent.

4. Summary
Magnesium oxide was studied using ramp wave compression with the Omega laser. Stepped targets of [100] MgO with 4 different thicknesses were fabricated using lithographic techniques. Free surface velocities across all thicknesses were measured with a VISAR. The measured wave profiles show the presence of an initial, weak MgO elastic precursor, followed by a shock to ~60 GPa due to the elastic precursor of the diamond ablator, and then ramp compression to maximum stress as high as 234 GPa. The peak elastic stresses ranged from 3-5.5 GPa for our ~25-40-μm thick samples. The pressure-density equation of state of MgO under ramp loading lies between previous static and shock studies, consistent with the expectation that modest heating (~1000 K) is achieved in these experiments. Our results are also consistent with recent X-ray diffraction measurements on ramp-loaded MgO.

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