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SESAME EQUATION OF STATE NUMBER 7611, BERYLLIUM OXIDE

by

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ABSTRACT

A new equation of state (EOS) for beryllium oxide (BeO) has been constructed for the SESAME library as material number 7611. Unlike the existing EOS for BeO in the library (7610), this new EOS incorporates the effect of a structural phase transition which has been predicted by two independent theoretical calculations.

Beryllium oxide (BeO) is of considerable technological importance due to its use as a moderator in nuclear reactors. It is also a material which has been of recurring interest to various users of the SESAME library. For these reasons, it is important to ensure that the SESAME library contains the best possible equation of state (EOS) for BeO at all times.

The task of generating an EOS for BeO using the computer program GRIZZLY[1] is complicated by a scarcity of empirical data to be used as input. In general, the most important data for such a calculation is Hugoniot data, since these data provide information about the EOS at high pressures. Unfortunately, the best available Hugoniot data for BeO are not amenable to an unambiguous interpretation (see Fig. 1).[2] When the current EOS for BeO on the library (material number 7610) was generated, this difficulty was avoided by using models which only require empirical data at ambient conditions. Although the output Hugoniot for 7610 provides a reasonable straight-line fit to the data (in the form of shock velocity u_s vs. particle velocity u_p), the degree of scatter about that line is quite substantial (see Fig. 1).

We have generated a new EOS for BeO (material number 7611) by reinterpreting the existing Hugoniot data in the light of two fairly recent theoretical calculations on the phase stability of BeO at high pressures.[3,4] Both of these calculations predict a large volume discontinuity phase transition from the wurtzite structure to the rocksalt structure under pressure. The first of the two calculations predicted a transition pressure of 217 kbar using the pseudopotential method,[3] while the other calculation obtained a transition pressure of about 400 kbar using the potential-induced breathing (PIB) model.[4] Given the general quality of modern theoretical phase stability calculations (see for example Ref. 5), we would anticipate that the wurtzite to rocksalt phase transition in BeO is real and should occur at a pressure less than 1 Mbar.

In seeming contradiction of the theoretical results, hydrostatic data on BeO reveal no evidence of a phase transition for pressures up to 550 kbar.[4] However, for a large volume discontinuity structural phase transition involving a change in coordination number such as the predicted wurtzite to rocksalt transition, the wurtzite structure is likely to remain metastable for pressures well above the equilibrium transition pressure. Thus, although the hydrostatic data probably rule out the lower of the two theoretical predictions for the transition pressure, they are not inconsistent with the 400 kbar prediction.

Hugoniot data often reveal phase transitions which do not appear in hydrostatic data. Examination of the Hugoniot data for BeO in Fig. 1 reveals that the four highest pressure points can be fitted with a straight line which does NOT intercept the u_s axis at the sound speed. This is consistent with the assumption that those points are associated with a different structure than the initial one. In addition, the three points in the range $1.3 < u_p < 2.0$ km/s show a distinct flattening which is characteristic of the mixed phase region for a structural phase transition. We have therefore fitted the Hugoniot data with a 3 straight-line fit defined by the (u_p, u_s) points: (0.0, 8.5), (1.52, 10.48), (1.83, 10.48), and (3.00, 12.22). (See Fig. 1.) That fit to the data implies a transition pressure of 479 kbar, in reasonable agreement with Ref. 4.

For 7611, the compressed cold curve (zero temperature isotherm) was obtained for compressions up to 1.5 by removing the thermal contributions from the Hugoniot specified by the 3 straight-line fit in Fig. 1. For higher compressions, the cold curve was required to smoothly merge with a cold curve obtained by mixing the TFD cold curves for the two constituents.[1,6] In the expanded region, the cold curve was required to have a Lenard-Jones form with FACLJ = 0.3 (Ref. 1) and with a cohesive energy of 141 kcal/mole.[7] This part of the calculation with GRIZZLY also required the reference density for BeO (3.008 gm/cc).[2]

The thermal electronic part of 7611 was calculated with GRIZZLY by first using the TFD model[1,6] to generate the electronic EOS for each constituent (Be and O) separately and then obtaining the total thermal electronic contribution to the EOS of BeO via additive volume mixing.[1] The nuclear contribution to 7611 was calculated using the CHART-JD model.[8] These two components of the EOS required several additional items of empirical data: the atomic masses of the constituents (Be - 9.012, O - 15.999),[9] the Debye temperature (1280 K),[3] the reference Gruneisen γ (1.25),[2] and the melting temperature (2530 K).[7]

This new EOS for BeO is somewhat speculative, and additional theoretical work on the structural phase stability is clearly necessary. In the region above the proposed phase transition, 7611 does a significantly better job of fitting the empirical Hugoniot data than does 7610. However, the lower two segments of the 3 straight-line fit used here are questionable given the absence of any usable Hugoniot data for $u_p < 1.3$ km/s. Users of the SESAME library should be aware of these caveats when doing calculations involving BeO.

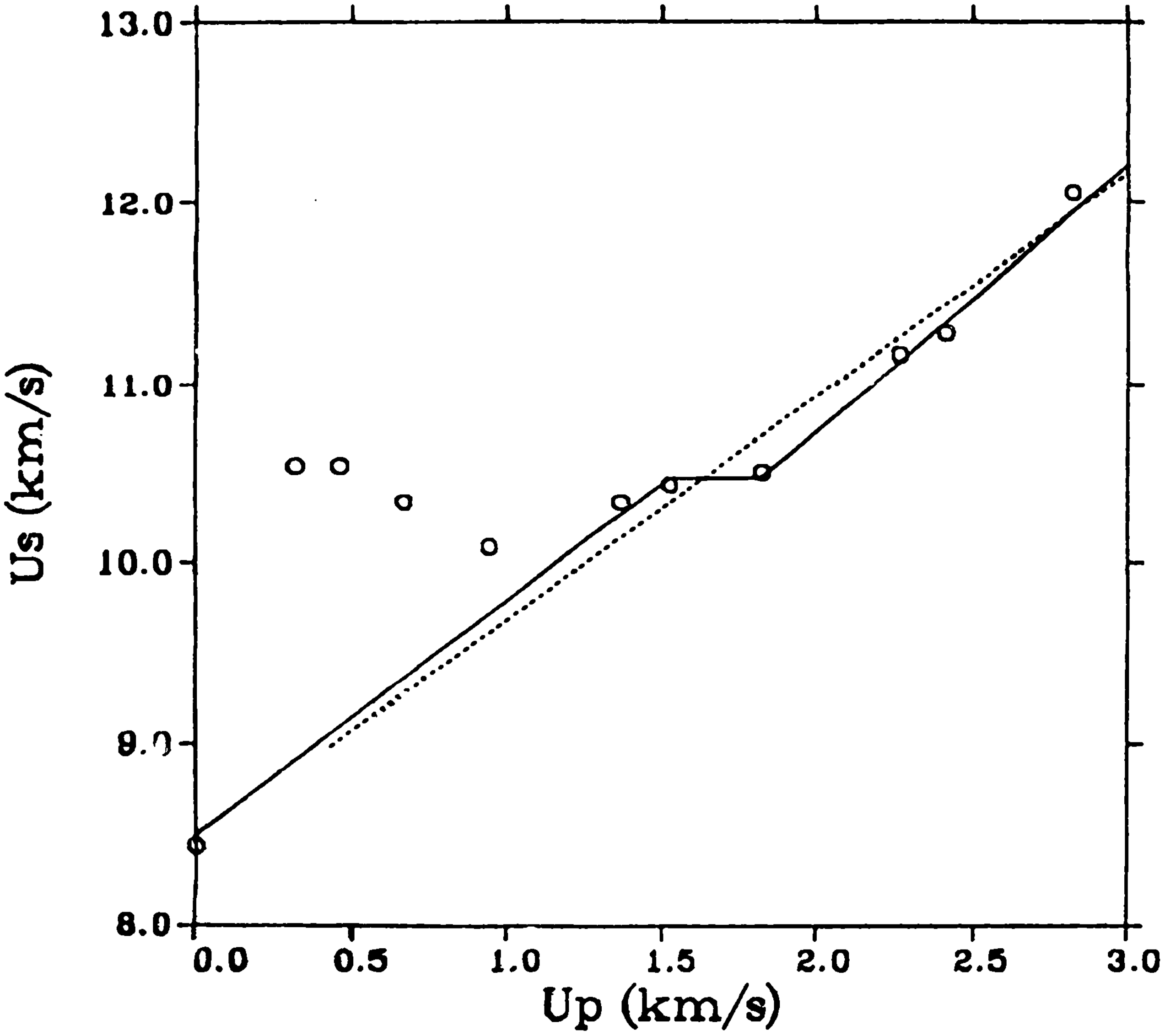

Figure 1. Calculated Hugoniots for material numbers 7611 (solid line) and 7610 (dashed line) compared with experimental data from Ref. 2.

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