

06 Similarity between shock-induced polymorphic transitions in the silica system

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A set of experimental data on the thermodynamic parameters of polymorphic transitions in the silica system is considered. Analysis of these parameters in dimensionless form is performed. A fundamental result of the analysis is that the thermodynamic parameters of all silica polymorphs after transitions are described by a single universal Hugoniot of polymorphic transition. It is shown that the two-shock model of polymorphic transformations proposed earlier by the author describes all the results obtained in the analysis. A joint consideration of the experimental data and model calculations leads to the conclusion that during a polymorphic transformation, the density of the new phase is determined from the condition that the elastic pressure components after the first shock and after the polymorphic transition are equal.

Keywords: silica, quartz, coesite, stishovite, fused quartz, quartz ceramic, polymorphic transformation, similarity, Hugoniot.

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Introduction

The propagation of a shock wave through a crystalline substance can lead to a polymorphic transition i.e. rearrangement of the atomic structure of the substance. The formation of new crystalline modifications in short time intervals represents one of the most interesting subjects in the shock wave physics and the high-pressure physics.

The researchers have continuously paid interest to shock loading of silica for more than half a century. In the geophysical applications, it is caused by the fact that SiO₂ is a dominant component in the Earth group planets. For physicists, as this system has several metastable polymorphs, it is a suitable model system for studying fundamental physical properties of the substance, in particular, polymorphic phase transformations. The present paper gives no chance of reviewing the results of the previous studies. Combined with the time transformation of the study fields, these reviews can also be found, for example, in [1–4]. This will only discuss results directly related to the problems reviewed in the present paper.

The earlier studies can be used to conclude that there is still no attempt of thermodynamic analysis of the known experimental results for the polymorphic transformation for the entire silica system as a whole. These results are plotted on Fig. 1 on the plane ($u_s - u_p$), where u_s — the shock velocity, and u_p — the mass velocity of a loaded substance behind the wave front both before and after the phase transition. These data are taken from [4–10] (note that they all, except for the data [4], are included in the database [11]). It should be noted that we consider only non-porous materials or materials with very low porosity of at most 3%.

It is clear from Fig. 1 that for each crystal substance the Hugoniot consists of several portions. The first portion corresponds to the Hugoniot of the first shock and is described, as usually, by a straight line: $U_s = a + \lambda u_p$, where a and λ — the empirical coefficients (for the quartz plot, it is illustrated by the experimental points and the straight line, which is extrapolated to the bigger values u_s ; the values a and λ used here are given in the table below). This section ends at some velocity $u_s^{(1)}$. Then, almost at the same value of the wave velocity, the mass velocity of the substance jumps to start a portion of the Hugoniot of the high-pressure phase. At the same time, as shown in [4] for quartz, the fully new phase is still unformed; it will

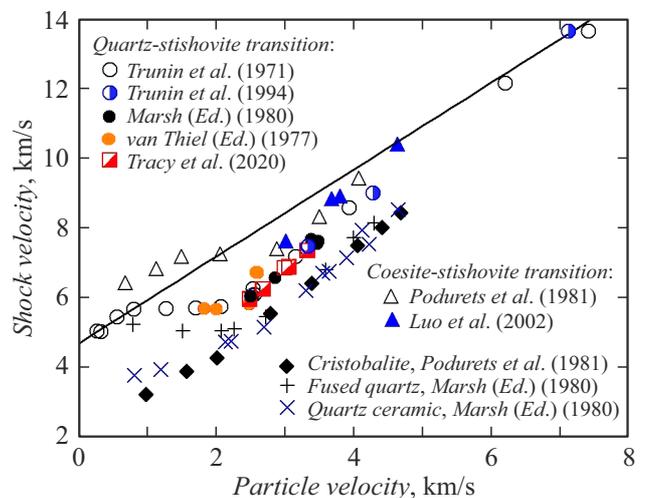


Figure 1. Data of the Hugoniot for the silica phase transition. The straight line is the adiabat of the first wave in quartz.

be later at some value $u_s^{(2)}$ of the wave velocity (it is not so obvious on the plot), and to be continued then by the proper Hugoniot of the new phase.

It also follows from the analysis of all the previous studies that until recently there was still no success in creating the thermodynamic model of the process, which would plausibly describe dynamic characteristics of the material after the phase transition for any crystal substance. This two-wave model based on the fundamental thermodynamic values was recently considered in [12–15]. As this model is used in the present paper, its main provisions should be mentioned.

— Parameters a and λ of the shock adiabat of the first shock are determined by linear approximation of the experimental data u_s and u_p near the beginning of the phase transition. Possible problems arising in case of a small number of the experimental points are in sufficient detail discussed in [12–15].

— The equation of the state of any phase is written in the Mi–Gr-neisen, wherein „cold“ (elastic) components of pressure and energy are taken as

$$p_e = \frac{K_0}{n} \left(\left(\frac{\rho}{\rho_0} \right)^n - 1 \right), \quad (1)$$

$$E_e = \frac{K_0}{n\rho_0} \left\{ \frac{1}{n-1} \left[\left(\frac{\rho}{\rho_0} \right)^{n-1} - 1 \right] + \left(\frac{\rho_0}{\rho} - 1 \right) \right\}, \quad (2)$$

where K_0 — the bulk compression modulus, $n = K'_0$ — its pressure derivative at $p = 0$, ρ_0 — the density in the normal conditions, and the index „e“ designates elastic components of pressure and energy. Note that like in [16], the small difference between the isothermal and adiabatic modules is not taken into account in the present paper.

— The analysis (2) shows that for all the phase transitions the elastic energy of the new phase decreases [12–15].

— The polymorphic transition is in the „phase jump“ — in the shock occurring directly behind the first shock front and being immobile in relation to the first shock front. The jump intensity reduces with increase in the shock velocity.

— In the phase transformation, the substance is additionally compressed due to reduction of the elastic energy.

— The density of the new phase at any shock velocity is determined from a condition of equality of the elastic components of pressure after the first shock and after the phase jump. Taking into account (1), we obtain

$$\rho_2 = \rho_{02} \left\{ \frac{K_{01}n_2}{K_{02}n_1} \left[\left(\frac{\rho_1}{\rho_{01}} \right)^{n_1} - 1 \right] + 1 \right\}^{\frac{1}{n_2}}, \quad (3)$$

where the indices 1 and 2 refer to the first and second phase, respectively.

— The other substance characteristics (pressure, mass velocity and internal energy) are determined from the classic equations of the dynamic compatibility at the shock front and the phase jump. In particular, for the pressure we obtain:

$$p_2 = p_1 + \rho_{01}u_s(u_{p2} - u_{p1}) = \rho_{01}u_s u_{p2}. \quad (4)$$

— The substance intersecting the shock front into the new phase fully transits into the new phase at the shock velocity $u_s^{(2)}$, and this transition is determined by the condition

$$|\Delta E_{e21}(u_s)| \geq k p_1(u_s) \left(\frac{1}{\rho_1(u_s)} - \frac{1}{\rho_2(u_s)} \right), \quad (5)$$

where ΔE_{e21} — the change of the specific elastic energy of the substance, and k — the empirical coefficient. The comparison with the experimental results for various substances, including those outside the silica system, has provided that $k \approx 1/2$.

— The model does not describe the non-equilibrium state of the substance and that is why, generally speaking, it is fair for $u_s \geq u_s^{(2)}$, and at the same time it does not take into account possible effects of melting or evaporating of the substance. For some substances (for example, graphite and boron nitride) $u_s^{(1)} \approx u_s^{(2)}$.

The values of the parameters characterizing the substances considered in the present paper are given in the table. The accepted densities for the substances considered in the paper are taken from [11], wherein the value of the cristobalite density given in [9,11] specifies that the experiments used porous samples with the porosity of $m \approx 1.08$ (the ratio of the densities of the solid and porous materials). Although, it is stated above that only the results for porous samples with $m \leq 1.03$ will be considered, here, an exclusion was made due to lack of other results. The calculation evaluations for the model in this case were made similar to [15]. For the cristobalite, [16] specifies the value of the bulk compression modulus $K_0 = 11.5$ GPa. In our case, the estimated value of 10 GPa was taken, as [15] demonstrated that for obtaining plausible results of calculations for the porous material the value K_0 had to be reduced. For the quartz ceramics, due to no data for K_0 , the estimated value was also taken.

For the number of the substances (quartz, coesite, stishovite, fused quartz), the literature specifies many evaluations of the bulk compression modulus in the initial conditions. The present paper evaluated the values K_0 and n based on the condition of the best description of the known experimental results for the normal isotherms by means of the equation (1). Note that values for K_0 obtained and given in the table are within the ranges of the values thereof for the corresponding substances (which are known from the literature) and, in particular, they are close to those specified in [16]. Like in [17], the bulk velocity of sound in the normal conditions has been evaluated from the relationship $\rho_0 c_{0B}^2 = K_0$.

When prior to the beginning of the phase transition there is a sufficient number of the experimental data for u_s and u_p , the parameters a and λ of the shock adiabat of the first shock can be determined by the least square method. Otherwise, the procedure for determining these parameters is described in detail in [12–15].

Values of the characteristic parameters of the substances accepted in the present paper

Substance	ρ_0 , g/cc	K_0 , GPa	n	c_{0B} , km/s	a , km/s	λ	$u_s^{(1)}$, km/s	$u_s^{(2)}$, km/s
Cristobalite	2.13	~ 10	3.5	2.17	1.932	1.391	3.3	—
Quartz	2.65	37–39	4	3.84	4.757	1.25	5.8	7.2
Coesite	2.92	105	3.5	6.0	2.808	1.389	6.5	8.7
Stishovite	4.31	300	5	8.34	—	—	—	—
Fused quartz	2.2	37	—	4.19	—	—	5.1	—
Ceramic quartz	2.15	11.5	—	2.3	—	—	1.6–2	—

1. Results of investigation

The sections of the experimental dependences of Fig. 1 referring to the phase transition adiabat specify the possible existence of a similarity between the phase transition process for the said substances. Let us consider the experimental results in dimensionless form. For each substance we will take the velocities in relation to the bulk speed c_{0B} of sound, so will the pressures in relation to $\rho_0 c_{0B}^2$. Thus, for the shock velocity, the mass velocity and the pressure, the dimensionless values are introduced: $U_s = u_s/c_{0B}$, $U_p = u_p/c_{0B}$, $\Pi = p/K_0$. As a result of this replacement, the data for the Hugoniot presented above on Fig. 1 transform to the form shown on Fig. 2. In contrast to the first plot, the shock velocity and the mass velocity of the substance have been reversed on Fig. 2. In the author’s opinion, it is more logic as the experiment and the calculations specify the shock velocity exactly, and the desired values are characteristics of the substance after the phase transformation. It also includes the calculated dependence $U_p(U_s)$ for quartz. Figure 3 shows the experimental data for the Hugoniot after the phase transition as the dependence $\Pi(U_s)$ together with the calculation result as per the model for the quartz-stishovite transition.

The procedure for determining the calculation values required for comparing with the experimental data is quite simple. The standard equations of the dynamic compatibility are written at the fronts of the first shock and the phase jump to be supplemented by the formulas (1)–(3). The obtained system of equation is used to find the required values in dependence on the shock velocity.

2. Discussion

The data of Fig. 2 confirm the above-said about the possibility of existence of similarity between the processes of polymorphic transformation in the silica system: all the experimental dimensionless data for all the considered crystal substances of the system are laid along one curve, which with the increase in U_s converges, in particular, to the Hugoniot of the first shock in quartz. At the same time, prior to the beginning of the phase transition

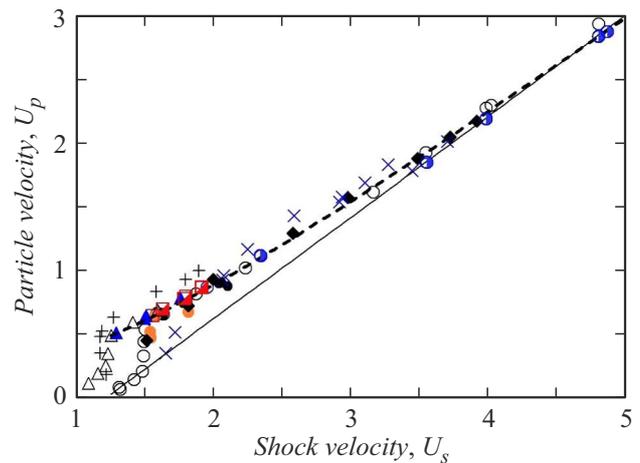


Figure 2. Dimensionless data for the Hugoniot for the silica phase transition. The straight line — the extrapolated adiabat of the first wave in quartz, the discontinuous line — the calculation as per the polymorphic quartz-stishovite transition model being considered in the present article. The experimental data are designated in the same way as on Fig. 1.

($u_s \leq u_s^{(1)}$) each substance has its own shock adiabat both dimensionally, and dimensionlessly. For the non-crystal substances of the system there is the same trend of converging to the quartz Hugoniot.

Figure 3, which represents the data for the dependence $\Pi(U_s)$ of the pressure on the shock velocity, also demonstrates the available similarity of the polymorphic transitions for all the considered substances of the system. The similar pattern is also observed for the experimental dependences $\Pi(U_p)$ and $U_s(U_p)$. Thus, we have obtained the universal set of the Hugoniot of the polymorphic transition for the silica system. This set is of a fundamental character, as it allows determining the dynamic characteristics of any crystal substances of the system at the polymorphic transformation in dependence on the shock velocity. Note that in case of the dependences $\Pi(U_s)$ (Fig. 3) and $\Pi(U_p)$, the experimental data for the amorphous substances of the system (fused quartz, ceramics) not only converge to the universal adiabats of the phase transition, but together with

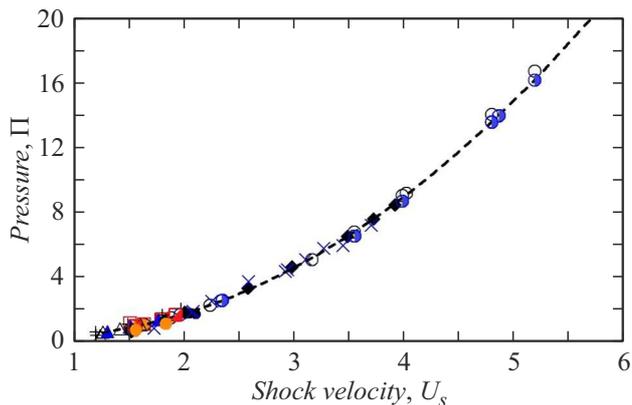


Figure 3. Dimensionless data of the Hugoniot for the silica phase transition. The discontinuous line — the calculation of $\Pi(U_s)$ as per the polymorphic quartz–stishovite transition model being considered in the present paper. The experimental data are designated in the same way as on Fig. 1.

the crystal substances they extremely accurate correspond to these dependences for all the values of the shock velocity.

Now, let us consider, in what extent the calculations as per the above-said thermodynamic model correspond to the results obtained. This model is selected, particularly, by the fact that [12–15] has specified well compliance of this model with the experimental data, in particular, it was demonstrated that it is the only one which fairly accurately describes the experimental dependences $u_p(u_s)$ after the phase transition for the various substances.

Previously, there were many efforts to build the models describing the kinetics of the phase transition in the conditions of the non-stationary process. The present paper gives no chance of reviewing these papers. We note only one fundamental moment. They were generalized in that in the vicinity of the beginning of the phase transition ($u_s \approx u_s^{(1)}$) there was postulation of an equation, which was useful to specify, in one way or another, a portion of the substance transferred into the new phase (for example, in [1,18] and in some other later papers). None of these models has not been generally accepted, and a cause thereof is partially explained by a critical remark of the author [18]: the above-mentioned „equation does not relate the transition way to the fundamental thermodynamic values, it does not specify any new notion on the basic mechanism of transition“. In addition to it, these models described only the transition itself, but they did not described the characteristics of the state of the substance after the transition at any shock velocity.

In our model the non-stationary section of the polymorphic transformation is not considered, but instead the expression (3) is introduced, whose physical meaning is described above and which allows determining the density of the new phase for any shock velocity. The data of the Figs. 2 and 3 illustrate that the design shock adiabat of the phase quartz–stishovite transition describes the experimental

results for the universal Hugoniot. The results of the calculations for the phase transitions coesite–stishovite and cristobalite–quartz have fully confirmed the conclusions, which are provided above for the transition quartz–stishovite. These results for $(U_s - U_p)$ — the Hugoniot are shown on Fig. 4. It is clear from the plot that all the Hugoniots of the phase transition (the curves 1–3) are almost the same. We note that it is possible to achieve their ideal coincidence, for example, by small correction of the values a and λ for the first shock. Therefore, it can be concluded that the model under consideration describes the above-noted experimental result, i.e. the existence of the universal Hugoniot of the phase transition for the silica system substances. It is clear from Fig. 4 that for all the substances, the Hugoniots of the first shock, with increase in its velocity, converge to the universal adiabat of the phase transition and merge into it at some point. More exactly, the shock adiabat of the phase transition intersects them at a small angle. The physical meaning of this intersection for each substance means that the intensity of the phase jump becomes zero (the zero change of the characteristics of the substance at its front), the phase jump is lost and, further on, the phase transition and the state of the substance are determined by the Hugoniot of the first shock. Figure 4 illustrates it with the experimental data which are not shown on the Figs. 1 and 2: these data [6] for quartz at the shock velocities $u_s = 20$ km/s (the pressure after the phase transition $p \approx 630$ GPa) and at $u_s = 33$ km/s ($p \approx 1.9$ TPa). It is clear that these points are on the Hugoniot of the first shock for quartz (the line 4). We remind once again that the design line I of the phase transformation shown here for high values U_s for clarity

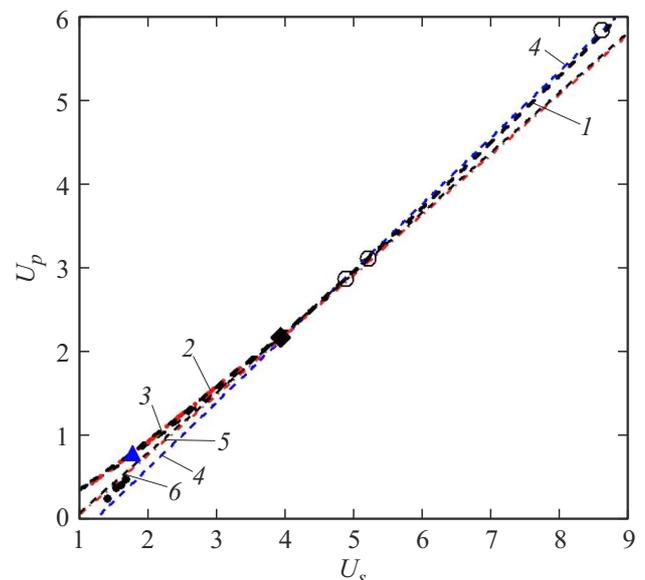


Figure 4. Results of the calculations $(U_s - U_p)$ — of the Hugoniots of the phase transition (I — quartz, 2 — coesite, 3 — cristobalite) and the first shock (4 — quartz, 5 — coesite, 6 — cristobalite) in comparison with the experimental data. The black circles mean stishovite, while the other designations of the experimental data are the same as on Fig. 1.

and passing on the plot a little below the line 4, does not physically exist already. For the other substances, for which the calculation has been performed, there are no experimental data with the high values U_s ; while for the two substances Fig. 4 shows the points of the databases with the maximum values of the shock velocity. Besides, for comparison, Fig. 4 shows the experimental data [19] for the stishovite Hugoniot.

Note that all the above does not contradict a traditional the two-wave diagram of the phase transition (see, for example [20,21]), and the difference from it is just causes of appearance of the phase jump and determination of the time when there is transition from the two-wave configuration to the single-wave configuration.

Taking into account the performed comparison of the design and experimental data, the above conclusion on the existence of the universal adiabat of the phase transition should be supplemented with the fact that the universal set of the Hugoniot of the polymorphic transition for the silica system exists at the stage of the process, when there is the two-wave configuration.

Now, let us consider other results which are obtained in the calculations using the model [12–15] of the polymorphic transformation.

We have selected the bulk speed of sound of each substance in the normal conditions as a main determining parameter when passing to the dimensionless values. Let us consider what will be with the above-obtained results, if the used value of this speed is not quite correct. In this case, both the experimental and model-calculated points remain on the corresponding universal Hugoniot, shifting upwards or downwards along it. The similar situation will be observed with the design points, if the model assumes that the phase jump is not immobile in relation to the first shock, but has some velocity in relation to it. It turns out that the only option when the calculation curve „leave“ the experimental universal Hugoniot is to reject the expression (3) in the model. Thus, we have actually obtained the confirmation of one of the assumptions laid in the model [12–15]: at the polymorphic transition the density of the new phase is determined based on equality of the elastic components of the phase pressure after the first shock and after the phase jump.

Besides, it is possible to note another moment positively characterizing the model under consideration. The [4], which has performed *in situ* studies of the crystal structure of the shock-compressed α -quartz, has noted that at the pressure of ~ 63 GPa the α -quartz peaks disappeared, thereby indicating the full transition into the crystal phase of the high pressure. In the model under consideration, the substance fully transits into the new phase at the velocity $u_s^{(2)}$ determined by the expression (5) with the equation mark. For quartz, it occurs at $u_s^{(2)} \approx 7.2$ km/s and at the same time the phase jump pressure is ~ 60 GPa.

Now, let us focus on the fact that the present paper does not discuss the density of the substance, more exactly,

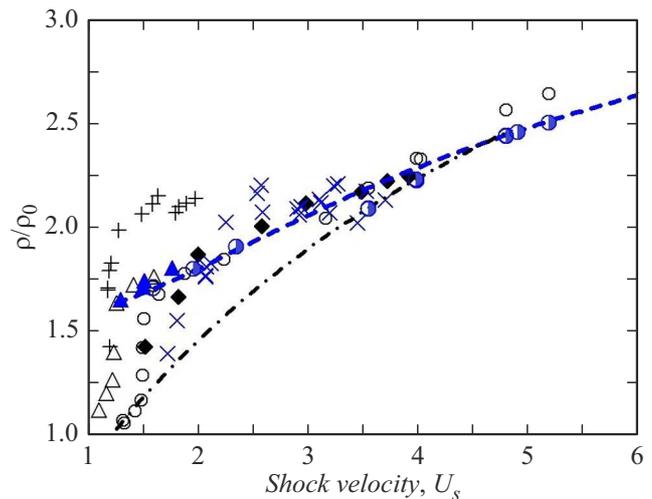


Figure 5. Dependence of the substance compression degree of the silica system on the dimensionless velocity of the shock. The dots — the experimental data, the lines — the calculation as per the model for the transition quartz-stishovite. The dash-dotted line — the Hugoniot of the first shock, the dashed line — the Hugoniot of the polymorphic transition.

the dimensionless value of the substance compression degree $\sigma = \frac{\rho}{\rho_0} = \frac{V_0}{V}$ (V — the specific volume) after the polymorphic transformation.

Figure 5 shows the results for $\sigma(U_s)$. It can be immediately noted that here we have not such ideality, with which the design Hugoniot of the polymorphic transition of quartz described the experimental data for all the substances of the Figs. 2 and 3. We remind that the „experimental“ data for the pressure and density in all the databases are obtained by means of test-measured velocities u_s and u_p by calculating from the equation of mass and pulse conservation, and at the same time the data „dispersion“ is somewhat increasing. If we do not pay attention to it, then it can be mentioned that the design Hugoniot of polymorphic transition in quartz quite well describes the results for the crystal substances of the system (it is true, however, that for coesite there is the trend to position the experimental results a little above the design curve), but poorly describes the results for non-crystal substances. The similar conclusions can be made for the dependence $\Pi(\sigma)$, too, whose results are shown on Fig. 6. Here, in comparison with the previous plots, it also included the experimental and design data [22] for fused quartz.

Based on the results of the Figs. 5 and 6, it can be concluded that the Hugoniot using the density of the substances are described by the universal dependences, apparently, only in case of crystal substances of the silicon system.

Now, let us return to the above-discussed most important universal dependences, which describe all the substances of the silica system. The design curves for quartz, which described the experimental data after the polymorphic

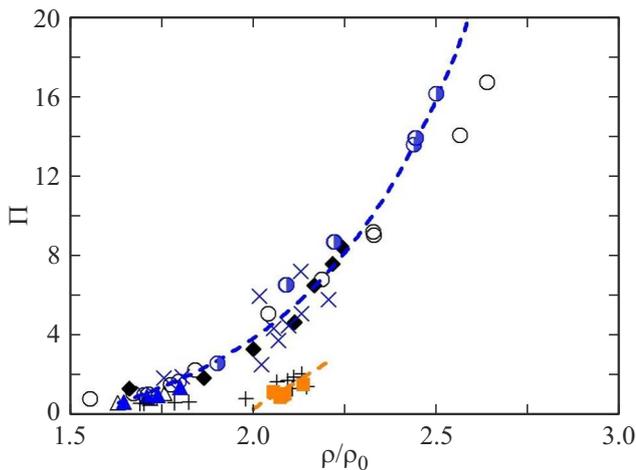


Figure 6. Data of the Hugoniot of the polymorphic transition for the dimensionless pressure $\Pi(\sigma)$. The experimental data are designated in the same way as on Fig. 1, the orange squares — the experimental data for fused quartz from the paper [22]. The blue dashed design curve is related to quartz (the present paper), so is the orange dashed one to fused quartz [22].

transformation of the Figs. 2 and 3, can be approximated by the below-presented analytical dependences:

$$\Pi(U_s) = 0.605 - 0.946U_s + 0.761U_s^2,$$

$$\Pi(U_p) = -0.249 + 1.116U_p + 1.331U_p^2,$$

$$U_s(U_p) = -0.131 + 1.332\sqrt{U_p} + 0.951U_p,$$

$$U_p(U_s) = \left[\sqrt{\left(\frac{1.332}{1.902}\right)^2 + \frac{U_s + 0.131}{0.951}} - \frac{1.332}{1.951} \right]^2.$$

The last two approximations can be substituted by more simple expressions which derive from the first two by means of (4):

$$U_p(U_s) = \Pi(U_s)/U_s, \quad U_s(U_p) = \Pi(U_p)/U_p.$$

All the given expressions ideally describe the design and experimental data up to $U_s \sim 8$, which, for example, for quartz, corresponds to the shock velocity $u_s \sim 30$ km/s.

At the end of the present study, we should mention another moment. There are quite substantiated assumptions that the universal Hugoniot of the polymorphic transformation obtained in this paper refer not only to the silica system, but they are also true for other substances in the conditions of the polymorphic transformation. The preliminary check has demonstrated that the experimental data for the polymorphic transition, which are used in [12] (graphite), in [14] (BN), as well as the data [23] (Si_3N_4), exactly correspond to the above-obtained universal Hugoniot. The final conclusion on the truthfulness of the specified assumption can be made after performing the additional studies.

Conclusion

The paper has analyzed many experimental data of shock-wave loading and the polymorphic transformation of the substances of the silica system. The analysis has been performed based on the thermodynamic characteristics of the process and avoided issues related to the change of the crystal structure of the substance. Based on the thermodynamics of the process of the polymorphic transformation, we have made it possible to detect new fundamental properties of the substances.

— It was found that the polymorphic transitions of the system are partially similar to each other. In the transition, the main dimensionless characteristics (velocities, pressure) at the process stage of the implemented two-wave configuration are described by the universal Hugoniot of the polymorphic transformation, which are the same for all the substances of the system. But, all the substances had their own Hugoniot of the first shock. It provided data indicating that this result can be exhibited in a wider class of the substances than the silica system.

— It has been demonstrated that the previously proposed model of polymorphic transformation describes a number of specific moments of the process. At the same time, one of the basic assumptions of the model was confirmed — the density of the new phase is determined based on equality of the elastic pressure components at the first shock wave and at the phase jump. Apparently, it is a fundamental property of the polymorphic transformation of the substance in the shock.

Conflict of interest

The author declares that he has no conflict of interest.

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