

# Dramatic effects of stress on metamorphic reactions

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## ABSTRACT

Thermodynamic calculations predict mineralogy from temperature and pressure and vice versa. Such calculations assume that stress is isotropic despite the fact that differential stresses prevail in Earth, resulting from large-scale tectonics and/or differences between fluid and rock pressures in porous rocks. New calculations show that differential stress can have significant effects on thresholds for metamorphic reactions, depending on the grain-scale reaction pathways. A differential stress may, depending on the reaction pathway, have an effect equivalent to a pressure difference on the order of (assemblage volume)/(reaction volume change) × (differential stress). The multiplying factor is typically 10 or more. For example, the onset of a garnet + clinopyroxene breakdown reaction may be offset, up or down, by the equivalent of 500 MPa in pressure for a 50 MPa differential stress. The effect is equivalent to a temperature difference on the order of (assemblage volume)/(reaction entropy change) × (differential stress). For example, the onset of muscovite + quartz breakdown may be offset, up or down, by the equivalent of 130 °C for a 50 MPa differential stress. Much of Earth is under differential stress, so the new calculations invite a reappraisal of metamorphic mineralogy and microstructure, indicating that new insights into stresses and fluid pressures on Earth can be gained.

## INTRODUCTION

Much of Earth is under differential stress, imposed at large scales through plate boundary forces and at the grain scale in porous media where fluid pressure differs from confining pressure. Imposed stress will persist during metamorphic reactions, but its influence on them is poorly understood. This is because the main geological applications of thermodynamics assume systems under isotropic stress (same force per unit area in all directions), and experiments to determine mineral equilibria generally impose that. When differential stress (defined as  $\Delta\sigma = \sigma_1 - \sigma_3$ , where  $\sigma_1$  and  $\sigma_3$  are the maximum and minimum principal stresses) is present, it is generally assumed (e.g., Stüwe and Sandiford, 1994) that the mean stress ( $(\sigma_1 + \sigma_2 + \sigma_3)/3$ ) equates to the numerical pressure relevant for thermodynamic calculations. There is a wide range of differential stress estimates in Earth (see the GSA Data Repository<sup>1</sup>); here 50 MPa is used for illustration. If one principal stress is reduced by this value, the mean stress will change by only 17 MPa; this is small in relation to the likely errors on pressure estimates. It is commonly assumed from such order-of-magnitude calculations that differential stress has a modest effect (although see Stüwe and Sandiford, 1994). Instead, it is shown here that this mean stress is not an adequate description of the effects of differential stress, and that those effects may be much larger than previously an-

icipated. The conceptual model also provides a basis for understanding coupled deformation and reaction.

## TWO THEORIES OF METAMORPHISM

There are two established theories unified here. First, in standard thermodynamics, under isotropic stress, the driving force for reaction is often written as  $\Delta G = G^{\text{products}} - G^{\text{reactants}}$  (where  $G$  is Gibbs free energy), so reaction may proceed if  $\Delta G < 0$  (or if the affinity,  $A = -\Delta G > 0$ ; Pattison and Tinkham, 2009). In this paper, for simplicity, stoichiometric mineral formulae are assumed. The Gibbs free energy,  $G$ , then equates to chemical potential,  $\mu$ , and the condition for products to form can be written as

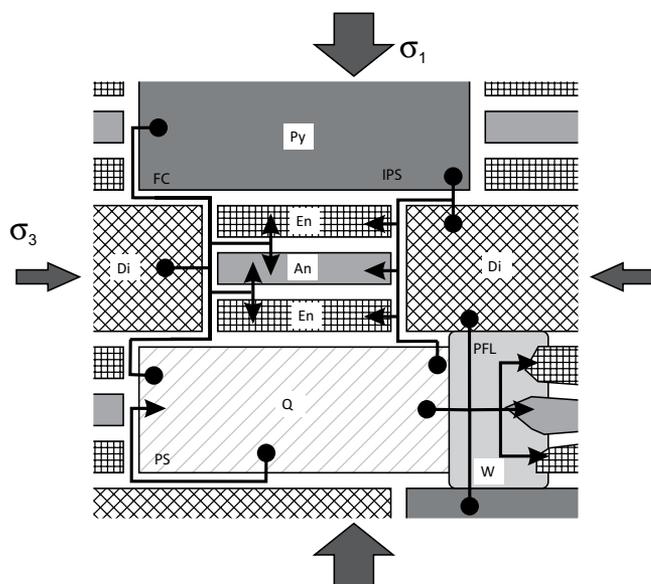
$$\mu^{\text{products}} - \mu^{\text{reactants}} = \Delta\mu < 0. \quad (1)$$

The second theory concerns diffusion driven by differential stress. Microstructures provide evidence that a single mineral can dissolve from interfaces with high normal stress and precipitate at interfaces with low normal stress: this is pressure solution (Rutter, 1976) or diffusion creep. It is explained via established theory for chemical effects in stressed systems (Gibbs, 1906; McLellan, 1980). The Gibbs free energy of a mineral is not defined under stress (Kamb, 1961); instead, individual interfaces under different normal stress have different chemical potentials:

$$\mu = F + \sigma_n V \cong \mu(\sigma_n), \quad (2)$$

where  $\sigma_n$  is the normal stress across an interface (compression positive; Pa),  $F$  is the molar Helmholtz free energy of the solid (J/mol),  $V$  is the molar volume of the solid (m<sup>3</sup>/mol), and  $\mu$  is the chemical potential of the stressed solid (J/mol) (Gibbs, 1906; McLellan, 1980). The notation  $\mu(\sigma_n)$  in the approximation denotes the functional dependence of the chemical potential of a phase on an isotropic pressure equal to normal stress  $\sigma_n$  (temperature dependence being implicit). Equation 2 shows that interfaces with higher normal stress,  $\sigma_1$ , will have higher local chemical potentials and higher local chemical concentrations than those under  $\sigma_3$ . Consequently, diffusion occurs, leading to dissolution of grains at  $\sigma_1$  interfaces and precipitation at  $\sigma_3$  interfaces: individual grains, and hence the entire rock, change shape. Figure 1 shows the reaction pathway PS for pressure solution: a pathway

**Figure 1. Model microstructure indicating various reaction pathways. Dots indicate dissolution at particular interfaces; arrowheads indicate precipitation. IPS—incongruent pressure solution pathway; FC—force of crystallization pathway; PFL—fluid pressure pathway; PS—pressure solution pathway;  $\sigma$ —stress; Py—pyrope, Di—diopside, Q—quartz, En—enstatite, An—anorthite; w—water in a pore. Gaps between grains are schematic grain boundaries—fast diffusion pathways (relative to intracrystalline diffusion).**



<sup>1</sup>GSA Data Repository item 2014241, a compilation of some differential stress estimates, a justification for the approximation in Equation 2, and the general version of the theory illustrated here, is available online at [www.geosociety.org/pubs/ft2014.htm](http://www.geosociety.org/pubs/ft2014.htm), or on request from [editing@geosociety.org](mailto:editing@geosociety.org) or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

is defined here as the transport route between the interfaces at which phases dissolve (dots in Fig. 1) and interfaces at which they precipitate (arrowheads).

These two theories are both well established: the first addresses reactions involving multiple minerals under isotropic stress, and the second addresses the chemical response of a single mineral to differential stress. Here these hitherto separate theories are brought together. In pressure solution, a particular mineral dissolves and reprecipitates elsewhere, but a more general scenario involves dissolution of several minerals with precipitation of new minerals. This is sometimes referred to as incongruent pressure solution (Fry, 1982), and microstructures offer clear evidence for it in nature (Brodie, 1995) and in experiments (Rutter et al., 1985). Despite such observations, it is generally implicitly assumed that the differential stress (which was present during reaction) will not have a significant effect on the mineralogy of the rock, although it obviously affects the microstructure. Although reactions may or may not proceed depending on kinetic factors, thermodynamics tells us a reaction is feasible if its affinity  $A > 0$ , where

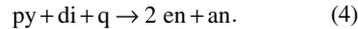
$$A = -\Delta\mu = \sum_{j=1}^N c_j \mu_j \quad (3)$$

Here  $c_j$  and  $\mu_j$  are the coefficients ( $>0$  for reactants) and chemical potentials of phases in a balanced reaction. Under isotropic stress, each phase has a unique value of  $\mu$ , but under differential stress the chemical potentials depend also on which interface is dissolving or growing during reaction (Equation 1): so, as expanded upon in the following, there are multiple reaction pathways and multiple affinities in stressed systems. I explore the consequences using example reactions (for the general theory, the Data Repository). I reiterate that, generally, the Gibbs free energy is not defined in a stressed system, and there cannot be any global equilibrium. Any preconceived idea that a specific mineral assemblage can be the most stable in a stressed rock must be abandoned. This general point has already been illustrated: if quartz is undergoing pressure solution, it is not even in chemical equilibrium with itself, and so there should be no expectation of a global equilibrium with other phases.

### EXAMPLE: PRESSURE-SENSITIVE REACTION IN METABASIC ROCK

This example is motivated by observations of metabasic rocks where garnet, clinopyroxene, and quartz dissolved at high-stress interfaces to form fibrous plagioclase and orthopyroxene on low-stress interfaces (Brodie, 1995). Garnet and clinopyroxene formed at 800–900 °C and 800–900 MPa. A second stage of metamorphism formed plagioclase and orthopyroxene

as conditions changed. For simplicity, Mg end-member compositions for the ferromagnesian minerals and pure Ca plagioclase are taken, so with standard abbreviations (Powell and Holland, 1988), we have



Under isotropic stress, the affinity will be

$$A = \mu^{py} + \mu^{di} + \mu^q - 2\mu^{en} - \mu^{an}. \quad (5)$$

If  $A > 0$ , the right side (enstatite and anorthite) is favored. The affinity does not dictate the details of reaction kinetics but its sign is a fundamental indication of whether a reaction can occur. At fixed temperature, if stress is isotropic then all the chemical potentials are a function of a single pressure  $P$  and, using Equation 1,

$$A = -\Delta F - P\Delta V, \quad (6)$$

where  $\Delta V$  is the net volume change of reaction indicating the sensitivity of affinity to pressure; in this case it is positive, so the right-side assemblage is favored at low pressures. This is a standard thermodynamic treatment.

With isotropic stress, all pathways have the same affinity. Which pathways are favored then depends on the kinetics of diffusion along grain boundaries and of attachment and detachment of chemicals at interfaces. With differential stress, different reaction pathways will be associated with different chemical potentials (Llana-Funez et al., 2012), and consequently each pathway has a different affinity. For example, suppose the reactants dissolve at interfaces

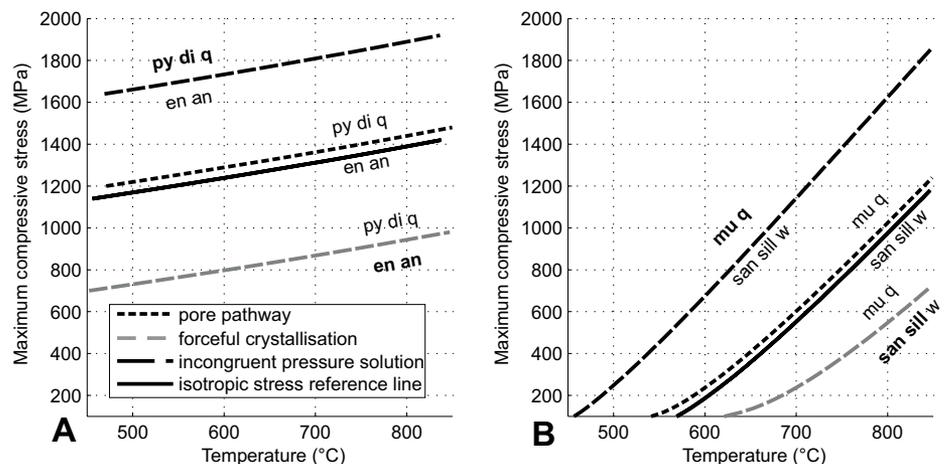
under high normal stress,  $\sigma_1$ , and the products precipitate at interfaces under low normal stress,  $\sigma_3$  (pathway IPS, incongruent pressure solution, in Fig. 1), then

$$A_{IPS} = \mu^{py}(\sigma_1) + \mu^{di}(\sigma_1) + \mu^q(\sigma_1) - 2\mu^{en}(\sigma_3) - \mu^{an}(\sigma_3). \quad (7)$$

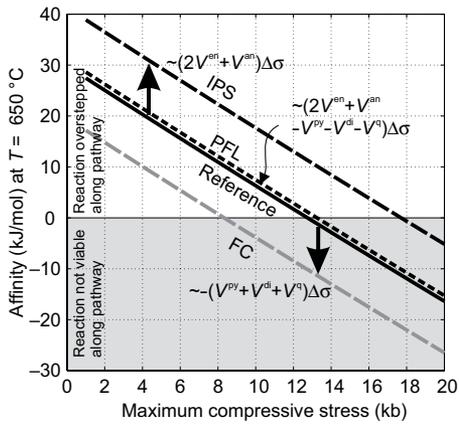
From which, using Equation 1, we have

$$A_{IPS} = -\Delta F + V^{py}\sigma_1 + V^{di}\sigma_1 + V^q\sigma_1 - 2V^{en}\sigma_3 - V^{an}\sigma_3 = -\Delta F - \Delta V\sigma_1 + (2V^{en} + V^{an})\Delta\sigma. \quad (8)$$

The value of  $A$  can be calculated using standard thermodynamic data (e.g., Powell and Holland, 1988) for any pair of values  $\sigma_1$  and  $\sigma_3$ . The  $A = 0$  line gives the threshold conditions for whether reaction is feasible on that particular pathway. To visualize that, first make stress isotropic ( $\sigma_1 = \sigma_3$ ) and calculate where  $A = 0$ ; this would be the equilibrium line for the reaction under isotropic stress (Fig. 1). Then suppose that  $\Delta\sigma = 50$  MPa, so now Equation 8 gives a different dependence of  $A$  on  $\sigma_1$ . Figure 2 shows  $A = 0$  depending on temperature,  $T$ , and  $\sigma_1$ ; the  $A_{IPS} = 0$  line is offset by ~500 MPa from the isotropic stress line. The  $A_{IPS} = 0$  line is not an equilibrium, but a threshold indicating an upper limit (in  $\sigma_1, T$  space) on whether reaction can occur along the IPS pathway. If  $\sigma_1$  is below that line, the reaction may (subject to kinetic factors) occur along the reaction pathway. If  $\sigma_1$  is above that line, the reaction cannot occur along that pathway, regardless of kinetics. The large offset of the  $A_{IPS} = 0$  line shows that differential stress has a dramatic effect on whether the reaction can occur. This is because the last



**Figure 2.** Lines of zero affinity ( $A = 0$ ) for various reaction pathways, for two reactions. For each line, mineral names in bold are those reacting at interfaces under  $\sigma_1$  ( $\sigma$ —stress; hence having high chemical potential) and those in light font are under lower normal stress  $\sigma_n$  (either solid-solid boundaries or solid-fluid boundaries). These are not equilibrium lines, but indicate the conditions at which reactions are thermodynamically feasible along the particular reaction pathway indicated. Reference line is for equilibrium under isotropic stress; pressure equal to  $\sigma_1$ . A: Garnet + clinopyroxene breakdown (py—pyrope, di—diopside, q—quartz, en—enstatite, an—anorthite). B: Muscovite + quartz breakdown (mu—muscovite, san—saniidine, sill—sillimanite, w—water).



**Figure 3. Affinity versus  $\sigma_1$  for different reaction pathways, for temperature  $T = 650$  °C. IPS—incongruent pressure solution pathway; FC—force of crystallization pathway; PFL—fluid pressure pathway; py—pyrope, di—diopside, q—quartz, en—enstatite, an—anorthite;  $V$ —volume. Reference line is for  $\sigma_1 = \sigma_3$ . For IPS and FC pathways,  $\sigma_3 = \sigma_1 - 50$  MPa (differential stress  $\Delta\sigma = 50$  MPa). For pathway PFL, fluid pressure =  $\sigma_1 - 50$  MPa. Because all the phases involved in reaction are solid, their volumes are roughly constant, so for each pathway the arrows and equations indicate approximate offset from reference value of affinity.**

term in parentheses in Equation 8 is the molar volume of the reaction products, a number (in this example)  $\sim 10\times$  larger than  $\Delta V$ . Hence, for this reaction pathway, there is a  $10\times$  greater dependence of affinity on  $\Delta\sigma$  than dependence on pressure in the case of isotropic stress. Figure 3 illustrates the values of affinity for different reaction pathways and dependencies of  $\sigma_3$  on  $\sigma_1$  (for  $T = 650$  °C).

Consider a second pathway in which dissolution occurs at low-stress interfaces and precipitation occurs at high-stress interfaces. This is akin to force of crystallization (FC), which is known to occur in experiments (Correns, 1949) and may occur in rocks (Carmichael, 1987) (Fig. 1). Then

$$A_{FC} = \mu^{py}(\sigma_3) + \mu^{di}(\sigma_3) + \mu^q(\sigma_3) - 2\mu^{en}(\sigma_1) - \mu^{an}(\sigma_1), \quad (9)$$

and the corresponding  $A_{FC} = 0$  line is shown in Figure 2. Because the reaction products are under high normal stress at the growing interfaces, they can grow only at much lower  $\sigma_1$  values;  $\sim 500$  MPa lower than the reference line.

A third pathway is transport via pores if fluid is present; the fluid need not necessarily participate in the reaction. The fluid pressure is predicted to be the sole control on affinity because all the growth is at interfaces under normal stress equal to the fluid pressure (path PFL in Fig. 2).

$$A_{PFL} = \mu^{py}(P_f) + \mu^{di}(P_f) + \mu^q(P_f) - 2\mu^{en}(P_f) - \mu^{an}(P_f). \quad (10)$$

This is numerically equal to  $-\Delta G$  under a pressure equal to fluid pressure,  $P_f$ . To illustrate, a fluid pressure equal to  $\sigma_3$  (for simplicity, ignoring mechanical consequences; Fig. 2A) shows a relatively minor (although still significant) effect: from Equations 1 and 10, we have

$$A_{PFL} = -\Delta F - \Delta VP_f = -\Delta F - \Delta V\sigma_1 + \Delta V(\sigma_1 - P_f), \quad (11)$$

and the sensitivity to changes in  $P_f$  is given by  $\Delta V$ , a smaller value than the scaling of  $\Delta\sigma$  in Equation 9.

These three pathways represent different possible general behaviors. In the specific example, formation of the reaction products was interpreted as linked to extension ( $\sigma_1$  vertical and due to the weight of overlying rock;  $\sigma_3$  horizontal), with  $\sigma_1$  decreasing through time. Reaction products were diagnosed as relating to a  $\sigma_1$  decrease (due to removal of overlying rock) of  $\sim 150$  MPa, using calculations based on isotropic stress (Brodie, 1995). In the new model, assuming the IPS pathway was in operation, that apparent decrease could be accounted for by imposing a horizontal tension of just  $\Delta\sigma = 15$  MPa, with no change in  $\sigma_1$  (from comparison of Equations 6 and 8). These calculations apply to all rocks under stress and undergoing reaction. The representative value of 50 MPa differential stress has an effect equivalent to an apparent pressure change of  $\pm 500$  MPa. Equating pressure to depth gives an apparent depth change of  $\pm 18$  km, a significant magnitude. Consequently, geodynamic models for metamorphic belts, which are built in part on apparent pressure estimates, may require reappraisal. A differential stress may, depending on the reaction pathway, have an effect equivalent to a pressure difference on the order of (assemblage volume)/(reaction volume change)  $\times$  (differential stress). The multiplying factor is typically  $\sim 10$  or more (Table DR2 in the Data Repository). In the following, I show that temperature estimates also require reappraisal.

### EXAMPLE: TEMPERATURE-SENSITIVE DEHYDRATION REACTION

Even if reactions have small molar volume changes, and are temperature sensitive (steep lines on  $P$ - $T$  grids), the theory given here is relevant. This is because it is the individual molar volumes of phases that come into the calculation, not just the overall volume change. Consider a simple dehydration reaction such as  $\mu + q \rightarrow \text{san} + \text{sill} + \text{water} (\text{w})$ .

Suppose that, in a stressed rock, muscovite and quartz dissolve at high-stress interfaces while the products precipitate from fluid. Then, muscovite and quartz will have higher chemical potentials than if they were breaking down in pores. The equation

$$A_{IPS} = \mu^{\text{mu}}(\sigma_1) + \mu^q(\sigma_1) - \mu^{\text{san}}(P_f) - \mu^{\text{sill}}(P_f) - \mu^{\text{w}}(P_f) \quad (12)$$

(Fig. 2B) shows the consequences of a 50 MPa difference between confining pressure and fluid pressure. For the IPS pathway, reaction is predicted to initiate 130 °C lower than expected, a significant difference. In general the effect is equivalent to a temperature difference on the order of (assemblage volume)/(reaction entropy change)  $\times$  (differential stress).

### DISCUSSION

Equation 1 is widely used to quantify and explain pressure solution (e.g., Rutter, 1976), but the interactions of multiple phases are not considered; its use is implicit in treatment of local stresses induced by reaction (e.g., Carmichael, 1987), but these will be localized in space and die out with time. Calculations have not previously been extended to the general case of pressure solution running in parallel with reaction, in the presence of a large-scale persistent imposed stress field. The prediction that modest differential stresses make a big difference to the driving force for metamorphic reactions means that reappraisal of metamorphic assemblages is required. For kinetic reasons more than one pathway may operate simultaneously, and the influence of different pathways will depend on, for example, diffusion coefficients along those pathways. Diffusion through fluid will be much faster than along interfaces, so one might suppose that the PFL pathway dominates; however, this cannot be the general case, given, for example, the reality of pressure solution, experiments showing force of crystallization, and the occurrence of reactions in stressed systems without fluid (e.g., in the mantle). All of those involve diffusion along stressed solid-solid interfaces. Aligned new minerals, commonly seen, usually result from growth under stress, so the new theory is relevant for diagnosing growth conditions as a function not just of  $P$  and  $T$ , but of differential stress.

How can large-scale patterns of mineral assemblages be evaluated in the context of this theory? Organized patterns of isograds are described from many areas (e.g., the classic map of Barrovian zones in Scotland; Atherton, 1977). If differential stress played a role in index mineral growth, then less-organized patterns might be expected, if stress varied throughout such regions in complicated ways. There are several possible explanations for the organized patterns we see.

1. In contact metamorphism, differential stress levels may be low, because deformation is often absent in aureoles (as evidenced by microstructures; e.g., Pattison and Tinkham, 2009), and an assumption of nearly isotropic stress is acceptable.

2. In regional metamorphism, stress levels may be high but vary systematically with position. Strength profiles in idealized models of the crust commonly show simple stress versus depth relationships; stress variations are not necessarily chaotic. Then the regular pattern of isograds would reflect smooth spatial (and/or temporal) variations in  $T$ ,  $P$ , and differential stress.

3. Stress levels may be low during mineral growth in regional metamorphism, despite the intense deformation seen in places, if metamorphosing rocks are weak. Levels of >100 MPa have been documented but may relate to sudden events in rather cold rocks (Table DR1), while in rocks above 400 °C, stresses are lower. In any case, inferred stress levels may not be contemporaneous with assemblage changes. The possibility of low stress levels (~10 MPa during regional metamorphism) should not be dismissed.

4. In a variant on this idea, it is common to see faceted porphyroblasts that do not seem to have been affected by deformation during their growth in rocks in which strain is partitioned on small scale (e.g., Bell et al., 1986). Stresses may then have been partitioned, the porphyroblasts growing under low differential stresses.

5. During prograde metamorphism of the crust, fluid is expected to be present. If the PFL pathway is favored for mineral growth, then the apparent pressure calculated from the assemblage will be the fluid pressure. The PFL pathway is plausible from a kinetic viewpoint because fluids, if present, will provide fast diffusion pathways. Deep within a low-permeability orogen, the fluid pressure might be close to the mean stress in value. Under this circumstance (though not in general), the mean stress, not far from the vertical stress in value, will control the assemblage and the familiar pressure versus depth interpretation would apply.

Point 5 is supported by some experiments (Llana-Funez et al., 2012) but it cannot be the entire story. First, if all chemical effects in a rock were mediated by fluid at a single pressure, then deformation by pressure solution would not occur (because this is a chemical consequence of the difference between  $\sigma_1$  and  $\sigma_3$ ). The fact that pressure solution does occur, often in parallel with reaction in regional metamorphism, high-

lights the operation of different reaction pathways under some circumstances; crucially, along grain boundaries supporting high normal stress. A second problem with point 5 is that it cannot apply to deep Earth metamorphism where fluid is likely to be absent. For example, I am not aware of any suggestion that the reaction ringwoodite  $\rightarrow$  periclase + perovskite is assisted by the presence of fluid. This occurs at the upper to lower mantle transition: there is likely to be differential stress during such reactions. The conceptual model presented here does not require the presence of fluid. Pathways such as IPS could occur along dry grain boundaries or even through lattices: the quantitative analysis still applies, because it depends only on the normal stresses at the positions of growth and dissolution.

This discussion is not intended to dismiss any of these points, but illustrates that targeted field, microstructural, experimental, and theoretical studies are required to establish where this analysis will modify current interpretations of metamorphic rocks. I repeat that all I have done here is to unite two theories (thermodynamics under isotropic stress and the theory of pressure solution) that are, separately, generally accepted.

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