

## Effect of solubility on the dihedral angles: Empirical law, physics, and thermodynamic treatment.

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Dihedral angles significantly affect the grain scale melt or aqueous fluid geometry and hence affect the macroscopic mechanical and transport properties of the partially molten rocks and rock + aqueous fluid systems in the Earth's interior. Measurements of the dihedral angles have been performed intensively for various systems. In these data, it can be recognized that dihedral angles of the binary eutectic systems tend to decrease with increasing solubility of the solid component into the liquid phase. This law, obtained empirically, is useful because it enables us to estimate the dihedral angles from the solubility data. For example, small solubility of the silicate component into aqueous fluids and large solubility into melts predict larger and smaller dihedral angles, respectively, which agrees well with the experimental data. The underlying physics of this empirical law is investigated.

Using a lattice-like model, we clarify the effects the liquid composition, temperature, and pressure have on the solid-liquid interfacial tension of the binary systems. It is shown that the non-ideality of the chemical bonds between the binary components, A and B, causes a strong compositional dependence of the solid-liquid interfacial tension; as the concentration of the solid component A increases in the liquid phase, the number of AA bond increases and the number of AB bond decreases at the solid-liquid interface, which decreases the interfacial tension. It is also shown that the actual observability of this effect changes with temperature relative to the characteristic temperature determined by the non-ideality of the interfacial bonds. In our model, the characteristic temperature is equal to the critical temperature of the regular solution. Most of partially molten systems are in or near the high temperature range, and the realization of this compositional effect explains well the empirical relation between the dihedral angles and the solubility. We consider that this can be the case for the rock + melt and rock + aqueous fluid systems, as well as alloys and organic systems. Previous models (Eustathopoulos et al., 1972, 1974) predicted a characteristic temperature much higher than that of our model; most systems are in the low temperature range and the effect of non-ideality is hardly observed. It might be possible for this model to predict the observations, but the physical mechanism may be much more difficult to understand.

Applied to the binary liquid-vapor interface, the Gibbs adsorption equation has successfully derived the concept of surface activity to explain the compositional dependence. Holness [1997] emphasized the practical importance of solubilities for predicting dihedral angles of binary solid-liquid systems and tried to understand this on the basis of surface activity. Clarifying whether the concept of surface activity can be immediately applied to the binary solid-liquid interface or not is another purpose of this study, which has not been investigated so far. By expressing the present results in the form of the Gibbs adsorption equation, it is shown that the surface activity defined by the relative adsorption is no more useful for understanding the compositional dependence of the binary solid-liquid interfacial tension. This point is quite different from the binary vapor-liquid interfacial tension but is similar to the binary liquid-liquid interfacial tension.