

## Effect of pressure on liquid-liquid phase separation of aqueous sulfate solution observed in fused silica capillary tubes at elevated temperatures

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Melt-melt, melt-fluid and liquid-vapor immiscibilities are common in magmatic and hydrothermal evolutions, while liquid-liquid phase separation (L-LPS) is rarely documented. Recently, we observed the L-LPS in some aqueous sulfate solutions (e.g.,  $\text{MgSO}_4$ ,  $\text{ZnSO}_4$ ,  $\text{Li}_2\text{SO}_4$ )<sup>[1-3]</sup>; the aqueous solution was separated into a sulfate-rich liquid phase and a sulfate-poor liquid phase, coexisted with a vapor phase. The L-LPS is characterized by a lower critical solution temperature, which is considered to be a macro-scale property of polymer solutions. *In situ* Raman spectroscopic analyses also suggested that complex ion associations are responsible for the appearance of the second liquid phase<sup>[1-3]</sup>. However, studies of the pressure effect on the L-LPS is still limited.

In this study, we used fused silica capillary tubes to construct optical cells and investigated the pressure effect on the occurrence of L-LPS in aqueous  $\text{CdSO}_4$  solutions at pressures ranging from 5 to 50 MPa. Results showed that the L-LPS temperature increases almost linearly with increasing pressure at  $\sim 0.5$  °C/MPa. Calculations showed that the product of the L-LPS temperature ( $T$ ) and the dielectric constant ( $D$ ) keeps approximately constant at each phase separation point. Since the  $D$  value of a solution is closely associated with fluid pressure and temperature, the current study also supports the proposition that L-LPS is mainly ascribed to the complex ion association between metal ions and sulfate. In addition, the results imply that L-LPS may occur in low-pressure hydrothermal ore-forming fluids during metal transportation and accumulation.

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