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Sulfur Speciation in Hydrothermal Fluids

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The sulfur compounds released by volcanic eruptions, generally believed to be in the form of SO₂ and H₂S, may cause global cooling of the atmosphere. However, several recent field and experimental studies suggested that under moderately oxidized conditions hexavalent sulfur species may coexist with SO₂ in magmatic fluids and may later be directly emitted at volcanic vents, which contradicts some thermodynamic predictions. We have investigated sulfur speciation in magmatic-hydrothermal fluids by loading different amounts of dilute sulfuric acid into a hydrothermal diamond-anvil cell and performing in situ Raman spectroscopy at temperatures up to 700°C. Upon heating SO₄²⁻ disappeared beyond 100°C, and SO₂ formed at >250 °C probably due to reduction by the rhenium or

iridium gasket. With high-fluid densities (such as >0.9 g/mL), the initial acid and air bubble homogenized into the liquid phase and most sulfur was present in the form of either HSO₄⁻ or H₂SO₄ (the rest being SO₂) within investigated *T-P* conditions (with pressures up to 10 kb). With low-fluid densities (such as <0.2 g/mL), the system homogenized into the vapor phase and molecular H₂SO₄ appeared to dominate (with pressures less than 1 kb). These observations strongly suggest that hexavalent sulfur is stabilized by hydration in magmatic fluids.

Key words: sulfur speciation, magmatic fluid, hydrothermal fluid, volcanic gas

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