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An Experimental Study of the Solubility of Liquid Mercury in Octane and Dodecane at Temperatures up to 200ºC

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Despite the common association of mercury ores with petroleum, little attention has been paid to the possible role of the organic phase in the transport of this element. Most researchers have assumed that mercury transport is via aqueous solutions and the gaseous phase, and that the presence of liquid hydrocarbons in the ore-forming system is purely coincidental. However, the documentation of relatively high concentrations of mercury in petroleum from mercury-bearing geothermal systems, together with experimental data at low temperature (up to 63ºC) demonstrating the relatively high solubility of mercury in a variety of alkanes (e.g. Clever and Iwamoto, 1987), suggest that the organic phase may be important in forming deposits of this metal. Fein and Williams-Jones (1997) recognized this possibility, and, extrapolating the available experimental data, showed that the maximum solubility of mercury in octane is 260 ppm at 150ºC. This value is at least two orders of magnitude greater than that estimated for aqueous liquids and is comparable to the proportion of mercury that can be transported in the gas phase. Owing to the fact that mercury deposits typically form in the temperature range 100-200ºC, and the need for high temperature experimental data, we have performed a series of experiments on the solubility of mercury in alkanes at temperatures of 100, 150 and 200ºC. The experiments were conducted in titanium autoclaves, and involved measuring the solubility of metallic mercury in n-octane and dodecane. Mercury solubility was determined using the mass-loss method. Before each run, a current of nitrogen was passed through the autoclave for 20 to 40 minutes to remove atmospheric gases. Autoclaves were loaded with a known mass of alkane and open pre-weighed quartz ampoules containing liquid mercury. Special care was taken to ensure contact between the liquid mercury and the alkane at the experimental conditions. After each run, the autoclave was air-cooled down to room temperature. The alkane was removed from the ampoule by dissolution in acetone, then the acetone was evaporated at 60ºC, and the ampoule was re-weighed. The losses in mass of mercury measured were attributed to its dissolution in the alkane. The solubility of liquid mercury in octane and dodecane is illustrated in Figure 1. The highest solubility measured for mercury was 1050 ppm (in dodecane, 200ºC). Our data support the conclusions of Clever and Iwamoto (1987) that there is a systematic change in Hg solubility with solvent and a near linear relationship between the logarithm of the mole fraction of Hg with reciprocal temperature. Recently, Bloom (2000) reported the results of a study of mercury speciation in petroleum at 25ºC, and showed that dissolved and particulate Hg dominate in the oils investigated. Given the linear correlation of mercury solubility vs. 1/T K in our study, it is reasonable to assume that mercury speciation at elevated temperatures does not differ significantly from that of 25ºC, and that dissolved mercury is generally present as Hg. Using this assumption, we have modeled our data using a Scaled Particle Theory (SPT) for non-aqueous solutions developed for non-polar solutes and solvents (Pierotti, 1976). To fit the data to SPT, the mercury solubilities were represented in the form of Henry's constants. The Lennard-Jones parameters for solvents were taken from Pierotti (1976) and Wilhelm and Battino (1971). The temperature dependencies of the effective hard sphere diameter for the solvent were taken from Wilhelm (1973). By fitting the preliminary experimental data to the SPT model, the following parameters for Hg (org) were evaluated: a 2^{298}(effective hard sphere diameter)=3 x10^{-8}(cm, (da $_2$/dT) = -5.3 x10^{-12} cm K^{-1}, ε $_2$/k = 250 K. These yielded values of mercury solubility in alkanes that are in good agreement with our experimental data for octane and dodecane, as well with data reported in the literature for hexane and heptane. Our study indicates that very high concentrations of mercury can dissolve in alkanes and that hydrocarbon liquids may be important agents in the transport of mercury in many ore-forming systems.

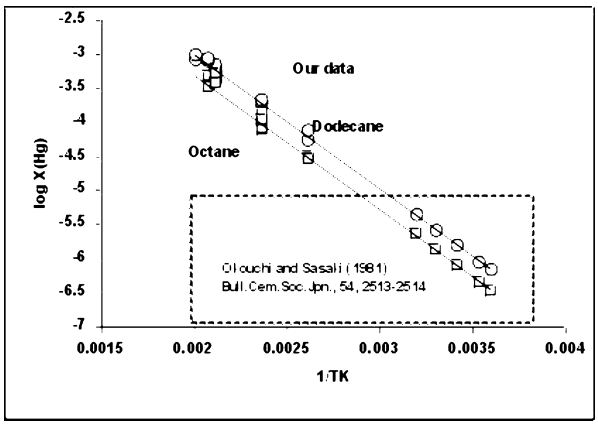

Figure 1

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