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Thermodynamic Modelling of Arsenic Behaviour in Subsurface Aqueous Systems

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Arsenic contamination is a serious problem at many contaminated sites and can pose a significant threat to human health and ecological systems. The aqueous speciation of As influences its solubility, transfer, and toxicity in subsurface waters. Although the fact that arsenic is relatively immobile in subsurface waters as a result of precipitation and adsorption onto iron hydroxides, it can present high concentrations in subsurface waters issued from various geological contexts (Figure 1).

Sources of As in the environment include geothermal springs, acid mine drainage, basin-fill deposits, and the application of various agricultural compounds (Rochette et al., 1999). The examination of arsenic contents (Figure 1) in various hydrothermal and subsurface waters shows that arsenic concentrations commonly exceed the SMCL imposed by WHO (World Health Organisation). In many contexts, these polluted waters are used for agricultural irrigation (rivers, lakes, etc.), therapeutic purposes (thermal sources), etc. Consequently, it is very important to know the behaviour of dissolved arsenic and understand the main mechanisms controlling its mobility and its dispersion in the environment.

In this study, the available literature data on the solubility of arsenic minerals in waters is critically reviewed (Gaskova et al., 1999). The existing conflicts between literature data is tentatively resolved for certain arsenic aqueous and mineral species. Surface complexation reactions seem to be the key mechanism for understanding the mobility of As, hence, thermodynamic stability constants for these solid-water interface reactions are also compiled.

This new thermodynamic database permitted the exploration of various hydro-geochemical systems ranging from oxidative weathering of mine tailings ($pH < 3.0$, $Eh > 500$ mV), through moderate conditions as in the case of soils (pH 6-8, Eh = 200 to 500 mV), to alkaline anoxic systems ($pH > 12$, $Eh < 0.0$ mV) as in the case of bottom ash leachates. The compiled thermochemical data is aimed at improving our knowledge and developing trapping process and remediation techniques. We have particularly examined the As behaviour in tailing leachates from the Cheni mine (France) that exploits gold from sulphide-quartz veins. High concentrations of As (up to 120 mg/l) were measured (Bodénan et al., 2000) in waters from the saturated zone. We have also explored the behaviour of As in alkaline fluids issued from bottom ash interaction with meteoric water (Freyssinet et al., 1998).

The most important results concern the following points. The critical review of existing thermodynamic data has permitted the enlargement of target solid phases for trapping arsenic excess in surface waters under various redox and pH conditions. Nevertheless, in view of the new SMCL (0.01 mg/l), solubility of arsenic minerals alone in surface and soil waters cannot provide As concentrations below this threshold. Other mechanisms potentially controlling As transfer processes (e.g., adsorption on oxyhydroxide surfaces, biological activity, etc.) are more attractive and are being used to develop remediation processes.

In the Cheni waters, redox equilibrium is not attained (Lassin and Azaroual, 2000). Consequently, the fate of arsenic during transport in the shallow aquifers could not be predicted by thermodynamic considerations alone. The redox disequilibrium and kinetic processes must be taken into account in numerical models. The preliminary PHREEQC (Parkhurst and Appelo, 1999) modelling results show that the transport of arsenic in shallow waters is controlled by adsorption-desorption reactions at water-oxyhydroxide solids.

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