

Approaching acid gas - fluid interactions

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Acid gases like CO₂ and H₂S as well as organo-sulfur compounds are common components in natural gas and oil reservoirs originating from a variety of sulfate reduction processes. Depending on the composition of a hydrocarbon reservoir, significant amounts of these objectionable compounds might have to be removed from the hydrocarbons during the refinery process.

Therefore, new disposal practices for acid gases and organo-sulfur compounds are needed due to the projected exploitation of giant hydrocarbon reservoirs with exceptionally high amounts of sulfur-containing compounds. One option for the disposal may be the geological sequestration within suitable strata. Although mixtures of H₂S/CO₂ have been injected into deep geological formations in Canada for more than 20 years,^[1] little is known about possibly occurring geochemical reactions in the underground.

In order to contribute to a better understanding of chemical processes under geological storage conditions, interactions of CO₂/H₂S, low-molecular organo-sulfur compounds and brines of different composition were investigated. Typically, experiments are conducted in sealed gold tubes at a temperature of 120°C and a pressure of 100 bar.

The experimental outcomes may provide a basis for a more detailed evaluation of the underground storage potential of organo-sulfur compounds and acid gases.

[1] Bachu & William (2004) *Geol. Soc. Special Publ.* **233**, 225-234.

Arsenic and old gold mines: mineralogy, speciation, and bioaccessibility

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Understanding the behavior of arsenic (As) during weathering of sulfide-bearing mineral deposits and associated mine wastes is important with regard to minimizing the exposure of humans and wildlife to this carcinogenic element. In low-sulfide gold-quartz vein deposits of the Mother Lode region of California, As occurs predominantly in the primary sulfide minerals arsenopyrite (FeAsS) and arsenian pyrite (Fe(S,As)₂). Detailed characterization of As- and iron- (Fe-) bearing primary and secondary minerals in mine waste piles at the Empire Mine State Historic Park (EMSHP), Grass Valley, California, was done as part of a larger study designed to evaluate the influence of mineralogy and speciation of As on bioaccessibility in mineralized environments and abandoned mines.

On the basis of electron microprobe analysis, arsenian pyrite at Empire mine has 0 to 5 wt. % As and arsenopyrite has 41 to 43 % As; secondary weathering products include hydrous Fe oxides (HFO, ferrihydrite and goethite, 0 to 18 wt. % As), hydrous Fe arsenates (HFA, including scorodite, FeAsO₄·2H₂O, and related phases, more than 20 wt. % As), As-bearing jarosite (KFe₃(SO₄,AsO₄)(OH)₆, 0.2 to 0.8 wt. % As) and unidentified Ca-Fe-arsenate minerals. The speciation of As in the secondary minerals is predominantly As(V) (at least 90%), based on x-ray absorption spectroscopy. The molar ratio As:Fe in HFO is about twice as high as that in arsenian pyrite grains in close spatial association; most likely, this reflects adsorption or coprecipitation of arsenate from solution on HFO. Based on electron microprobe analysis, HFO from Empire mine has a minimum of 1 wt.% SiO₂.

Bioaccessible As, measured using *in vitro* methods simulating conditions in the human stomach and small intestine, ranged from 1.4 % to 11% of total As in 25 samples of mine waste from the EMSHP area. In addition, a sequential extraction procedure [1] was applied to the 25 samples. The first two fractions (F1, 0.05 M ammonium sulfate; F2, 0.05 M ammonium phosphate) liberated sorbed As; the sum of F1+F2 concentrations was less than the *in vitro* test results for all samples. The third fraction (F3, 0.2 M oxalate at pH 3) liberated As associated with amorphous and poorly crystalline oxides of Fe and aluminum (Al). The sum of F1+F2+F3 concentrations was consistently higher than results from the *in vitro* tests, suggesting that some As associated with amorphous and poorly crystalline Fe and Al oxides is not bioaccessible. Adsorbed SiO₂ tends to lower the dissolution rate of HFO [2], reducing bioaccessibility.

[1] Wenzel *et al.* (2010) *Analytica Chimica Acta* **436**, 309-323.

[2] Eick *et al.* (2009) *Clays and Clay Minerals* **57**, 578-585.