

Chemical evolution of waters in long term interaction with granitic rocks: example of waters from Nagra boreholes, North Switzerland

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The kinetics of aluminosilicate water reactions has been studied mainly theoretically and extrapolated at very long time starting from experimental studies performed far from equilibrium. Some sets of waters such as those from Nagra boreholes, Northern Switzerland, allow to define some constraints on the times needed to reach equilibrium.

Waters obtained by Nagra in six boreholes in crystalline areas of the North Switzerland and waters of the granitic recharge area (Black Forest) were analyzed for both chemical and isotopic characteristics (Pearson *et al.*, 1991). When considered for several aspects, waters fall into four groups. These aspects are geographic location, water salinity, gases concentrations and isotopic properties:

1) the recharge group: these waters have undergone minimal water- rock interaction and have been very recently recharged.

2) the Eastern group present salinities, helium

content and isotopic characteristics consistent with recharge during the last glaciation.

3) the Western group: their chemical and isotopic properties indicate recharge conditions similar to present but predating the Eastern group: the recharge corresponds to the last interglacial period.

4) the saline group: their high salinity, large helium content, low C activity are consistent with the assumption of very old waters.

The chloride content (closely related to ^4He and ^{36}Cl concentrations) appears to be the best index for the extent of the water rock interaction.

The lack of precise data on pH and aluminum precludes a direct calculation of the departure from equilibrium. The silica content of waters is generally close to the chalcedony solubility at the formation temperature. Some waters from the saline group are at saturation with quartz.

The Na/K ratios of the water vary from a value of 2, close to the Na/K ratio in the rock, for

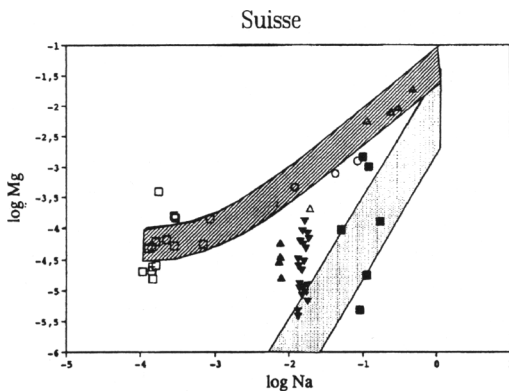


FIG. 1a. Mg concentrations versus Na concentrations in a log log scale

recharge waters
western group
sedimentary w.

eastern group
saline group
o mixed waters

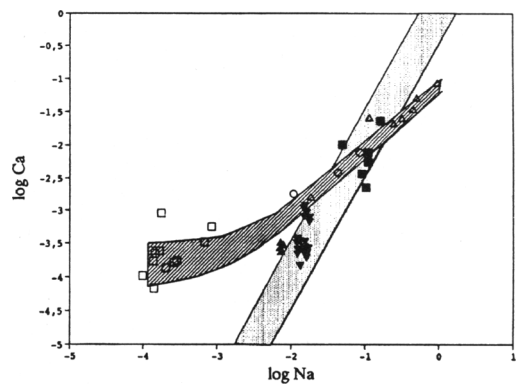


FIG. 1b. Ca concentrations versus Na concentrations in a log log scale

recharge waters
western group
sedimentary w.

eastern group
saline group
o mixed waters.

recharge waters to a value of 53 for waters from the saline group, a value close to the equilibrium value between albite and K-feldspar at the water temperature.

Calcium and magnesium were reported in log Ca (Mg) versus log Na plots. Na is highly correlated with Cl in these waters and can be used as an indicator of the extent of dissolution reaction. On the other hand the Ca(Mg)/Na² quantities at equilibrium are only slightly temperature and ionic strength dependent. The distinction between dissolution dominated waters (shaded area) and equilibrated waters (dotted area) is especially clear for magnesium (Fig.1). Recharge waters and waters from sedimentary rocks of the same area lie in the dissolution dominated field; waters of the saline group (essentially Weiach and Böttstein) fall in the equilibrium field. Almost all the other waters are intermediate between the two fields.

For calcium, all the borehole crystalline waters are in the equilibrium area.

Computer calculations (using PHREEQE code) confirm that Weiach waters are close to full equilibrium with quartz, albite, K-feldspar,

laumontite, prehnite, calcite and chlorite. Except the latter, all these minerals are prominent in the water bearing fractures of the crystalline basement.

Waters from Leuggern borehole present the same global chemistry although their temperatures vary from 25 to 66°C. Changes in potassium and magnesium content suggest that hotter waters are closer to equilibrium than the colder ones.

In summary, we can consider that the saline waters (about 100ky old) have reached full equilibrium. Waters of the eastern and western groups (some tens of ky old) present compositions which are within an order of magnitude consistent with equilibrium composition. The proximity to equilibrium is related to temperature.

References

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