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The ^{20}Ne/^{36}Ar Ratio as a Tracer for Ancient Oil: The Oil-Water and Gas-Water Double Distillation Model

Philippe Sarda (sarda@geol.u-psud.fr)[1], **Battani Anne** (anne.battani@ifp.fr)[2] &
Prinzhofer Alain (alain.prinzhofer@ifp.fr)[2]

[1] Dept. Sciences de la Terre, bât. 504, 91405 Orsay cedex, France
[2] Institut Français du Pétrole, 1-4, av. de Bois Préau, 92852 Rueil-Malmaison Cedex, France

Rare gas studies of either oil, natural gas or water from aquifers, have often reported fractionated rare gas abundance ratios. This is the case for the ^{20}Ne/^{36}Ar ratio where both ^{20}Ne and ^{36}Ar come from the atmosphere through equilibration with surface waters (Ballentine and O'Nions, 1994). In particular, the ^{20}Ne/^{36}Ar ratio measured in samples of natural gas (Ballentine et al., 1991) or water (Castro et al., 1998) may be as high as 1 or 1.5, and reaches the extremely high value of 5 in a recent study of natural gas in Mexico (Battani, 1999). This appears much enhanced compared to the expected values of 0.1-0.2 for air equilibrated with either sea water or fresh water at reasonable surface temperatures.

To explain such a fractionation in the case of natural gas, it has been proposed that gas had equilibrated with water at depth (Bosch and Mazor, 1988), which should result in rare gas "pumping" from water into the gas phase, where neon should be more efficiently transferred than argon because the solubility of argon in water is higher than that of neon. This would result in lowered ^{20}Ne/^{36}Ar ratios in the water. However, this process requires very small gas to water volume ratios of 10-3 or less, and it was shown to increase the ^{20}Ne/^{36}Ar ratio up to no more than about 0.6, given reasonable values for the initial ratio and temperature (Ballentine et al., 1991). Also, this model does not explain why elevated ^{20}Ne/^{36}Ar ratios are found in waters of some sedimentary basins.

For these reasons, we searched for another interpretation. In two recent rare gas studies of exploited natural gas wells in Pakistan and Mexico (Battani, 1999), where only gas-no oil-is known, we found that the high ^{20}Ne/^{36}Ar ratios appear together with the smallest ^{36}Ar concentrations. This suggests that the high ^{20}Ne/^{36}Ar ratios may be indicative of a residual phase which would have been depleted of its rare gases. Such a residual phase may be water, but rare gas depletion cannot be due to gas - water contact, which would induce a decrease of the ^{20}Ne/^{36}Ar ratios in the water phase.

We therefore propose that water was at contact with oil, in the past and at depth. This would actually induce a rare gas depletion and an increase of the ^{20}Ne/^{36}Ar ratio in the water, because argon and neon are more soluble in oil than in water, but oil has a stronger affinity for argon than for neon. This is confirmed by rare gas measurements in oil (Pinti and Marty, 1995). In this case, the ^{20}Ne/^{36}Ar ratio would be a tracer of ancient oil, which may have been lost from the basin as seems to be the case in Pakistan and perhaps in Mexico. The signature of the past presence of oil would thus be kept in water, and later transferred to gas.

We tested this idea using Henry's law to describe rare gas solubility in water and oil, and found that a Rayleigh distillation process is necessary to reproduce the elevated ^{20}Ne/^{36}Ar ratios and corresponding low ^{36}Ar concentrations. With such a Rayleigh distillation for both the oil - water and the following gas -water contact, we explain the data with temperatures of about 100-150 ºC for the oil-water distillation, and 20-60 ºC for the gas - water distillation. This simple model also allows to derive the oil to water volume ratio for the first stage, and the gas to water volume ratio for the second stage. Gas to oil volume ratios of 10-100 are thus derived for the present Pakistan and Mexico studies.

The double distillation model therefore permits to use the fractionated ^{20}Ne/^{36}Ar ratios to retrieve the relative quantities of oil that have been generated and lost in a sedimentary basin.

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