Production of radionuclides from gas reservoirs

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Recently, precipitates of native lead and galena in natural gas production installations have been reported to contain ^{210}Pb from the ^{238}U natural decay series. In these Naturally Occurring Radioactive Material (NORM) scales ^{210}Pb is frequently unsupported, i.e. there is no ingrowth from parent nuclides (Schmidt, 1998). Because of the half-life of ^{210}Pb (22.6 y), the origin of this radionuclide must be looked for in or near the respective gas reservoirs. Shales or coal measures are likely sources of ^{210}Pb in the North Sea area, but often uranium enrichments are also found in residues of migrated hydrocarbons. Therefore, a production core from northeastern Netherlands was studied for the presence of bitumens and naturally occurring radionuclides (NOR's) in Rotliegend gas reservoir sediments.

Sample description

All samples were taken from 4" cores of the Slochteren Sandstone section in the production core. The section consists of alternating conglomerates, coarse to fine sandstones, and shales, all indicative of a fluviatile deposition environment. Between 2800 and 2830 m, several intervals of brownish grey sandstone contain dark, bituminous layers, ranging in thickness from one mm to over 30 cm. The same intervals show a sometimes intense overprint of white anhydrite 'nodules'. In a preliminary γ-ray analysis, only the bituminous intervals showed anomalously high natural radioactivity. Samples were taken from three of the thickest bituminous layers for detailed investigation (Table 1).

TABLE 1

Sample	Depth (m)	^{238}U (Bq/g)	^{226}Ra (Bq/g)	^{210}Pb (Bq/g)
1	2811	2.2	2.3	2.4
2	2813	2.8	3.0	2.1
3	2827	2.8	3.0	2.9

Diagenesis

Before compaction, early diagenesis was characterized by the overgrowth of radial Mg-Fe-chlorite and/or illite on detrital quartz and feldspar grains. Upon burial, extensive pressure solution caused many sutured grain contacts. In a next stage, intense dissolution of quartz grains created abundant secondary porosity. Initially some Fe-Mg carbonate precipitated, followed by anhydrite, $CaSO_4$. Anhydrite cementation was most intense in the middle of the three bituminous sandstone layers and resulted in the formation of large anhydrite 'nodules'. The hypersaline $Ca\text{-}SO_4$-bearing brine which caused the precipitation of anhydrite probably originated from Zechstein sediments locally juxtaposed to the Rotliegend reservoir.

Shortly after, most of the the remaining pore space was filled with bitumen. Base metals such as Ni, Co, Cu, Sn and As, associating the oil, precipitated forming arsenides and sulphides on sediment grains and as inclusions within the bitumen. Sulphide was probably derived from the dissolution of anhydrite and subsequent reduction of SO_4^{2-} to S^{2-}. Ba was

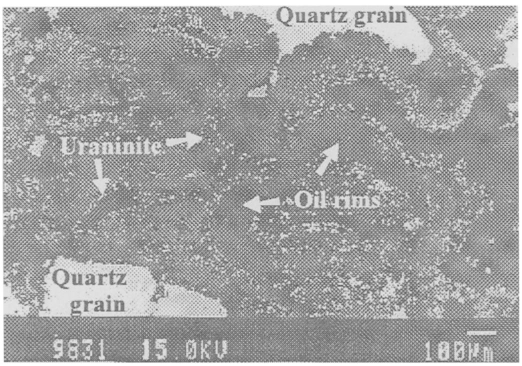

FIG. 1. Arrays of uraninite crystals along oil rims in a large secondary pore.

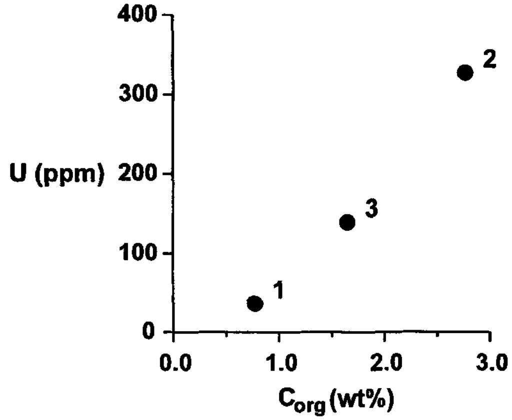

FIG. 2. U *vs.* C_{org} contents.

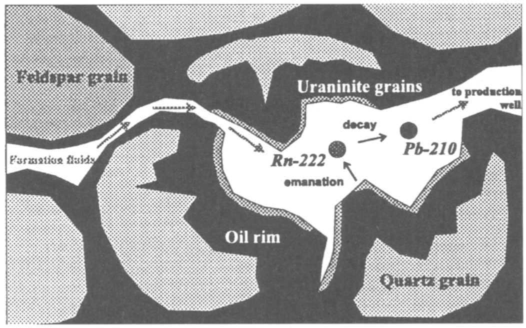

FIG. 3. Mobilisation of ^{210}Pb from uraninite through emanation of ^{222}Rn.

also carried into the sandstone and precipitated large crystals of baryte upon contact with anhydrite.

All bitumen contains U, yet only in the largest secondary pores, where quartz dissolution continued after oil entrapment, uraninite precipitated (Fig. 1). The correlation between U and C_{org} contents of the three samples (Fig. 2) suggests that U was already present in the oil when it became trapped in the sandstones. Chemical ages of 235 Ma, calculated from U and Pb contents of uraninite grains from sample 2, provide an age for the migration of the oil into the Slochteren Sandstone. The observed association of bitumen and anhydrite suggests that both fluids followed the same preferential migration path and probably originated from the same source. Therefore, a Zechstein source rock is most likely. Uranium and the other metals found in the bituminous sandstones were probably extracted from the Kupferschiefer.

Mobilisation of naturally occurring radionuclides

Radioactive daughter nuclides ^{226}Ra and ^{210}Pb are in secular equilibrium with ^{238}U in samples 1 and 3. In sample 2, ^{238}U and ^{226}Ra are in secular equilibrium, but ^{210}Pb shows a deficiency compared to ^{226}Ra (Table 1) after correction for ^{210}Pb decay between the dates of coring and γ-ray spectroscopy. This suggests that ^{210}Pb can effectively be removed from the host rock by percolating fluids. However, since all uraninite grains of sample 2 yield the same chemical age of 235 Ma, secular equilibrium in the sandstone must have been disturbed only very recently. Disturbance was probably caused by human activity. Production of natural gas from the studied reservoir started in the end of the nineteen sixties, generating large-scale fluid flow throughout the reservoir, including the area of the studied production well. ^{238}U daughter nuclides which enter sediment pores through the alpha-recoil process may thus migrate inside the reservoir (Fig. 3). Coproduction of chlorine-rich formation water with natural gas then causes the transport of ^{210}Pb into production installations, where it coprecipitates with normal Pb, forming NORM scales.

References

Schmidt, A.P. (1998) *J. Geochem. Expl.*, **62**, no. 1-3 [in press].