

# Radioactivity in deep geothermal heat and power plants of Germany

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## 1. INTRODUCTION

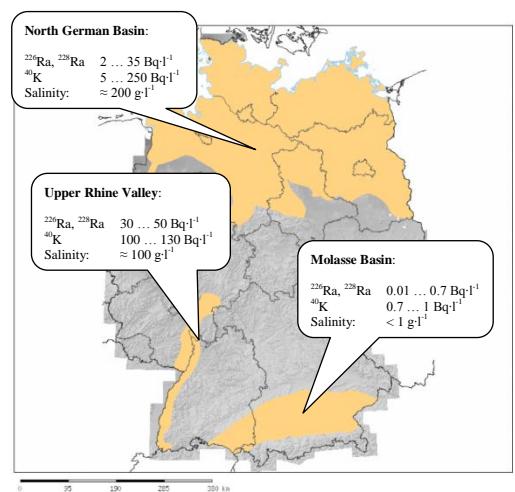
In Germany, deep geothermal heat and power plants use fluids from aquifers of different geochemical characteristics. The main regions with geothermal potential are: (i) the North German Basin (NGB) (II) the Upper Rhine Valley (URV) and (iii) the Molasse Basin (MB). Thermal waters from NGB and URV are highly mineralised (up to salinities of several  $100 \text{ g l}^{-1}$ ) in contrast to that from MB. Such highly saline fluids often carry enhanced concentrations of natural radionuclides which may cause the formation of scales with specific activities of several  $100 \text{ Bq g}^{-1}$ . Such materials are described by the term TENORM (Technologically Enhanced Naturally Occurring Radioactive Materials). Radiation protection measures are necessary for the safety of the employees. Residues must be disposed in compliance with radiation protection and waste management regulations. Geothermal energy production is thus added to the list of industrial practices involving TENORM in the draft of new EU basic safety standards [European Commission 2011].

During the last years a number of geothermal plants in Germany were investigated by the authors with respect to their fluid characteristics, scale properties and waste composition. Dose assessments for employees were carried out and ways of removing radioactive waste were tested and implemented. A comprehensive description of this work can be found in [Köhler et al. 2013]

## 2. RADIOACTIVITY IN GEOTHERMAL FLUIDS

The concentrations of natural radionuclides depend strongly on the geochemistry of the aquifer and of the hydrogeological parameters. Fig.1 summarises typical values of the activity concentrations as well as of the salinities. Highly saline brines as produced from hydrothermal aquifers in the North German Basin and in the Upper Rhine Valley are thus expected to contain enhanced radionuclide concentrations. Hereafter we will consider only geothermal plants working with such saline fluids.

Waters from the crystalline basement show also salinities  $> 100 \text{ g l}^{-1}$  below  $\approx 1000 \text{ m}$  depth [Bucher and Stober 2010]. So, petrothermal systems will also have to face the consequences of radioactivity in the fluid.



**Figure 1: Regions with geothermal potential in Germany [geotis] and their typical radionuclide concentrations and salinities.**

All investigated brines showed concentrations of U- and Th-isotopes below detection limit. The nuclide composition is specified by Radium isotopes of different half-lives:  $^{226}\text{Ra}$  (1600 y),  $^{228}\text{Ra}$  (5.75 y) and  $^{224}\text{Ra}$  (3.63 d) and by the radioactive Potassium isotope  $^{40}\text{K}$ .  $^{210}\text{Pb}$  was also found in several systems at some  $10 \text{ Bq l}^{-1}$ . A main source of the Radium content in thermal waters is alpha recoil at the solid/fluid interface of the aquifer rock. The  $^{40}\text{K}$  content reflects the Potassium concentration of several per cent due to the elevated salinity.

## 3. RADIOACTIVE PRECIPITATES IN GEOTHERMAL PLANTS

Production rates of saline brines in the order of  $100 \text{ m}^3 \text{ h}^{-1}$  will cause an annual activity throughput in the geothermal plant of  $10^{10} \text{ Bq}$  per radionuclide. Since changes of thermodynamic parameters as well as electrochemical processes lead to precipitation of substances from the fluid, a fraction of this activity permanently remain in plant components at surface.

Mineral phase analyses revealed similar composition both for scales from NGB and from URV: The first

phase consists of Ba/SrSO<sub>4</sub> mixed crystals with a minor fraction of CaSO<sub>4</sub>. A second phase is composed of mainly lead sulfides (NGB) or of mixed (Pb, Cu, Sb, As)-sulfides (URV).

Due to their similar chemical behaviour, Radium isotopes are effectively co-precipitated with Ba-containing sulfates and <sup>210</sup>Pb is deposited in the Pb-bearing sulfides. This process is associated with a radionuclide accumulation in the precipitates by four to five orders of magnitude compared to the fluid concentration. Typical specific activities of the formed scales are in the range of up to some 100 Bq g<sup>-1</sup> for <sup>226</sup>Ra, <sup>228</sup>Ra and in extreme cases of up to some 1000 Bq g<sup>-1</sup> for <sup>210</sup>Pb.

<sup>226</sup>Ra and <sup>228</sup>Ra specific activities are strongly correlated by their activity ratio in the brine. The absent correlation between <sup>210</sup>Pb and Ra-isotopes indicates independent formation processes (electrochemical reactions and pressure/temperature changes, respectively).

#### 4. RADIATION PROTECTION MEASURES

Annually generated scale masses of several 100 kg and their elevated specific activities produce a radiation field in the geothermal facility with equivalent dose rates up to some 10  $\mu$ Sv h<sup>-1</sup>. This status calls for the application of specific radiation protection measures in affected plants.

The usage of geothermal energy is presently not involved in the regulations of the German Radiological Protection Ordinance (Strahlenschutzverordnung, StrlSchV), neither in the specified fields of work with radioactive materials nor in the list of residues to consider. The situation will change within the next years in the course of the implementation of EU directives.

Nevertheless, even there is no obligation by the StrlSchV the operating company is responsible for the safety of their employees as well as of the unconcerned public and must react on the occurrence of enhanced radioactivity levels.

Radiation protection measures are necessary to be applied in the following fields:

- Operational radiation protection for the employees
- Removal of radioactive residues (waste) by deposition, combustion or melting

##### 4.1. Dose exposure of employees

For the estimation of the annual dose it is necessary to define the different pathways of radiation exposure during operation.

*External exposure* ( $E_{ext}$ ) will be caused by the influence of the gamma radiation field. This component is registered by measuring the distribution of the rate of the ambient dose equivalent, given in  $\text{Sv h}^{-1}$ . The contribution of the ubiquitous radiation

from radionuclides in soil etc. amounts to 60 ... 100  $\text{nSv h}^{-1}$ . Maximum dose rate values appear in a geothermal facility in the vicinity of heat exchangers and will reach at the surface of the components several 10  $\mu\text{Sv h}^{-1}$ . The consequence may be an order of a restricted access to such areas.

*Internal exposure* is the result of inhalation ( $E_{inh}$ ) or ingestion ( $E_{ing}$ ) of radioactive material. During the normal operation of a plant this component is negligible but working on opened plant components can lead to the uptake by men of dust particles containing  $\alpha$ - and  $\beta$ -emitters. Here exists an urgent need for instructions for occupational safety and for the handling of residues.

After an analysis of the situation, a radiological estimation and a compilation of realistic working scenarios it is possible for experts to estimate the mean additional annual dose for workers in the plant. A summary of the annual dose components as given exemplarily in Tab. 1 will be the result of such a survey.

**Table 1: Summary of annual dose components for employees in a geothermal plant (example).**

Typical scenario	$E_{ext}$	$E_{inh}$	$E_{ing}$	$E$
	mSv			
Stay in the plant	1,2	-	-	1,2
Filter replacement and maintenance	1,4	0,15	0,26	1,8
<b>Sum</b>				<b>3,0</b>

German regulations (StrlSchV) define a criterion of 6 mSv per year for the necessity of a notification of the works by the competent authority. In all investigated geothermal plants the annual dose for employees fell distinctly below this limit.

##### 4.2. Residues

Radioactive residues from a geothermal facility are of different types (Fig. 2.): pure scales, contaminated materials like filters, soil and cleaning tools and also metallic scrap. Some of them are combustible or can be recycled. Their specific activities range from  $< 1 \text{ Bq g}^{-1}$  to a few 1000 Bq g<sup>-1</sup>. All these materials must be removed according to the German StrlSchV in such a way that the additional annual dose exposure of the public falls below 1 mSv.



**Figure 2: Typical radioactive residues from a geothermal power plant: scales (left), filter (middle) and combustible byproducts (right)**

Materials exceeding  $0.2 \text{ Bq g}^{-1}$  must be removed according to the provisions of the StrlSchV. Furthermore, the acceptance criteria of the disposal sites as well as the transport law have to be fulfilled.

#### *Scales and similar materials*

Deposition of scales and scale-like materials at a landfill is possible, if the  $1 \text{ mSv}$  limit is not exceeded. This includes not only the exposure of workers but also that of the public by releases from the waste deposit. The critical pathway is the discharge by landfill leachate. Direct deposition is complicated for specific activities  $> 100 \text{ Bq g}^{-1}$  due to the expected high radionuclide concentrations in the seepage water. Immobilisation by so called Geopolymers improves the retention of radionuclides in the waste body. Realistic calculation scenarios on the base of leaching experiments are necessary to fulfil the administrative guidelines.

According to the acceptance conditions of the waste deposit certain chemical limits of harmful substances in the solid waste and in the leachate must be additionally fulfilled.

The described mode of disposal was tested and performed already several times with materials from geothermal plants and quantities at a scale of some tons.

#### *Combustible material*

German regulations require the combustion of flammable materials with a heating value  $> 6000 \text{ kJ kg}^{-1}$  like filter cartridges, contaminated overalls and foils in incineration plants. The enormous effort necessary for the dose assessment contrasts with the low waste amount in the order of some tons. Facilities for special waste burning therefore show a low motivation for the removal of contaminated material.

#### *Metallic scrap*

Metallic pipes, valves and other scrap are often contaminated at values below the monitoring limit of the StrlSchV. On the other hand, the acceptance levels of the recycling industry for radioactivity are more restrictive. The recycling by melting of TENORM contaminated metals in special recycling facilities is a suitable alternative and common practice in the removal of scale contaminated scraps.

## **5. CONCLUSIONS**

Radionuclide concentrations in highly saline fluids of German deep geothermal heat and power plants were found in the order of  $> 1 \text{ Bq l}^{-1}$ . Ra-isotopes and  $^{210}\text{Pb}$  can be accumulated in  $\text{Ba/SrSO}_4$ -and  $\text{PbS}$ -bearing precipitates, respectively, leading to the occurrence of scales with enhanced radioactivity levels ( $> 100 \text{ Bq g}^{-1}$ ).

The handling and the removal of such materials requires the consideration of appropriate radiation protection regulations.

Dose exposures of employees in geothermal plants are well below the annual limit of  $6 \text{ mSv}$ .

The disposal of residues depends on the type of material. Deposition of scales and similar materials and the recycling of metallic scraps have been already successfully executed several times.

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