ABSTRACT

Radioactivity of natural materials is widely studied mainly in connection to the nuclear fuel cycle. In the last decade the international radiation safety communities e.g. IAEA and Directorate General for Energy and Transport of the European Union paid huge attention to naturally occurring radioactive materials (NORM) used elsewhere i.e. in other industry. It was recognised that exposure of workers handling such materials can be very high, even substantially higher than occupational exposures of actual radiation workers. Being aware that a bulk amount of such materials are processed every day, the radiation safety community has published first safety standards related to NORM. But the natural radioactive materials are often used in research laboratories, educational institutions i.e. faculties as well as in museum collections, mostly in the form of geological specimens.

From 2002 onwards the Slovenian Nuclear Safety Administration (SNSA) became a responsible body for controlling radiation sources in industry and research as well as for controlling radioactive waste originating from all activities. An extensive campaign was systematically conducted in order to improve the inventory of radioactive sources. In 2007 the SNSA identified many radioactive sources in educational and research institutions dealing with geological samples. The samples containing uranium and thorium can have different forms e.g. ores, minerals and petrified samples. Many radiation safety requirements during handling and storing the samples were ignored because samples were not identified as radioactive materials or sources. Detected photon dose rates at the contact were a few orders of magnitude above the natural background and no provisions to avoid internal exposure due to radon were in place.

After the campaign of the SNSA inspections the further use of identified radioactive geological samples required the authorisation of such practice in line with all safety requirements published in Slovenian legislation, or the specimens were treated as a radioactive waste. The waste was taken over by the Agency for Radwaste Management and properly stored. The international guidelines related to practices concerning the radioactive geological samples would be beneficial for safe handling of such objects either in private or non private collections or institutes.

1 INTRODUCTION

In the nature around about 340 nuclides can be found, among them around 70 are radioactive [1]. Usually they are classified regarding their origin, namely primordial radioisotopes (e.g. $^{40}$K, $^{87}$Rb, $^{238}$U, $^{232}$Th), secondary radioisotopes and cosmogenic radioisotopes. Primordial radioisotopes have half-lives sufficiently long that they survived...
since their creation, i.e. comparable to the age of the universe, while secondary radioisotopes are the result of the decay of primordial radioisotopes. The cosmogenic radioisotopes are the result of the bombardment of stable nuclei by cosmic rays [2]. The military and non military activities of a man affected concentrations of some isotopes in the Earth. [3].

The exposure to radiation from radioisotopes either natural or man made depends on the activities performed, for example the characteristics of the exposure of aircrafts are very different than characteristics of occupational exposures when handling radioactive waste in a nuclear power plant. Some places in the world have levels of natural radioactivity well above average global values [1]:

- some mineral springs with water containing high concentration of $^{226}\text{Ra}$ and $^{222}\text{Rn}$
- some regions in Brazil, India, Iran and China with monazite sands or other radioactive mineral deposits
- areas of uranium ore deposits e.g. in Canada, Germany, Czech Republic, Africa, among them also a place of natural reactor at Oklo in Gabon.

Radioactivity of natural materials is widely studied in connection to the nuclear fuel cycle. In the last decade also the IAEA paid huge attention to naturally occurring radioactive materials (NORM) used in industries other than nuclear fuel industry e.g. use of phosphate fertilizers. It was recognised that doses to workers handling such materials can be very high, i.e. even substantially higher than occupational exposures of workers who are usually treated as occupational exposed workers. Being aware that a bulk amount of such materials are processed every day the radiation safety community has published first safety standards related to NORM [4, 5]. The European Comission also published recommendations regarding safe use of materials with enhanced natural occurring radioisotopes [6].

The radioactive materials are sometimes not used because their radioactive characteristics but due to other properties e.g. chemical or physical. For example the uranium and thorium chemicals found in chemical laboratories are very often not treated as radioactive materials so that no safety measures are present when handling such material or waste originating from activities in such laboratories [7]. Also among around more than 4000 minerals which are known today [8] some of them which are kept in laboratories or storages of institutes, faculties or museum collections are radioactive e.g. uraninite, pitchblende, brannerite, coffinite or thorite. Therefore they require special safety measures. The publications regarding safe handling of radioactive geological samples are quite limited.

2 RADIOACTIVITY OF GEOLOGICAL SAMPLES AND THEIR REGULATION

A few years ago the Slovenian Nuclear Safety Administration (SNSA) became a responsible body for controlling radiation sources in industry and research as well as for controlling radioactive waste originating from all activities. An extensive campaign was systematically conducted in order to improve the inventory of radioactive sources and establish regulatory control. In 2007 the SNSA identified many radioactive sources in educational and research institutions dealing with geological samples. The samples containing uranium and thorium can have different forms e.g. ores, minerals and petrified samples. Some samples inspected during the campaign are shown in Figure 1. The figure shows also the measurement instrument, namely radiation pager. The indication “9” shown on the pager indicates that the actual photon dose rate is above 38 µSv/h.
2.1 Identification of radioactive samples

The process of finding radioactive geological samples is similar to the process of finding other orphan sources described elsewhere [9]. When searching radioactive geological samples a well kept documentation regarding the origin of samples is very important. It can facilitate the identification of radioactive material among usually a large number of non radioactive samples. The detection of photons originating from radioactive material was performed by detection of external radiation using instruments like hand held gamma spectrometry unit *Fieldspec* shown in Figure 2.

Using such instruments the external photon radiation originating from thorium and uranium series [1] can be detected. The detected photon dose rates at the contact with samples are a few orders of magnitude above the natural background and no provisions to avoid internal exposure are in place. Besides the presence of uranium and thorium and theirs daughters other radioisotopes can be present in some samples e.g. coal samples contain also $^{40}$K.

The geological samples with enhanced radioactivity were stored in collections, laboratories, offices, halls and storages. Very seldom only one radioactive sample is present at a specific site, usually few tens of them were collected or exhibited together. The shapes of
specimens are very different as well as their homogeneity. The shielding of radioactive object by other objects can be a considerable problem in order to identify radioactive geological samples. At inspections all potentially radioactive samples were safely stored.

Later they were carefully investigated by qualified experts. Because of a huge amount of objects, namely few hundreds, the identification of radioactive specimens was very difficult. It was based on the exemption levels valid for $^{238}\text{U}$ and $^{232}\text{Th}$ given in EU directive [10] which are given in Table 1. The qualified expert developed simplified identification methodology by using measured dose rate, mass of an object and conversion factor between photon dose rate and activity concentration in order to compare the estimated activity concentration and appropriate exemption level. Details are given in [11]. According to data from [11] less than 5 % of potentially radioactive samples identified at inspections were later not identified as radioactive specimens.

As reported in [3] the global average values of $^{238}\text{U}$ and $^{232}\text{Th}$ in soil are about 30-50 Bq/kg. In specific soil and rocks the concentration can be orders of magnitude higher. In contrast to radioactive chemicals containing uranium and thorium the secondary equilibrium in geological samples is almost always present.

Table 1: Exemption levels for $^{238}\text{U}$ an $^{232}\text{Th}$ in secular equilibrium taken from [10]

<table>
<thead>
<tr>
<th>Series</th>
<th>Exemption levels</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}\text{U}_\text{sec}$</td>
<td>1 kBq/kg</td>
</tr>
<tr>
<td>$^{232}\text{Th}_\text{sec}$</td>
<td>1 kBq/kg</td>
</tr>
</tbody>
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### 2.2 Handling of geological samples

Many radiation safety requirements during handling and storing the samples were ignored because they were not identified as radioactive materials or sources. While using uranium and thorium radioactive chemicals in laboratories it is a good general laboratory practice in place providing that not substantial exposure to chemical and radiation toxicity exists, this is not the case when handling radioactive geological samples.

The risk associated with radioactive geological samples originates mainly from:
- external exposure due to decay of $^{238}\text{U}$ and $^{232}\text{Th}$ series
- internal exposure due to presence of long-lived radionuclides in a sample and radon with its short-lived decay products.

Both components should be assessed prior the use of geological samples. The assessment should be done for all lifetime phases of a use of radioactive samples i.e. from “its cradle to its grave” in order to avoid unjustified exposure of workers, students, visitors or private collectors. While prevention from external exposure can be achieved without huge expenses, the presence of accumulation of $^{222}\text{Rn}$ can be a major issue. In order to safely handle radioactive geological samples many measures can be applied, either administrative or physical, e.g.:
- use replica in museum collections and limit handling radioactive specimens as much as possible
- use ventilation before or during as appropriate when entering storages with radioactive materials
- use protective glass as for example lead glass to prevent exposure from gamma radiation
- use gloves and other personal protective belongings when handling radioactive samples
- use encapsulated specimens in order to prevent contamination
- use separate and suitable storages for radioactive geological samples
- define the specific area in laboratory where radioactive samples will be handled and control it
- establish written working procedure and maintain the records of samples.

Up to date no overview of safety measures in museums or institutes with radioactive geological samples was published.

Any practice with radioactive geological samples either storage, exhibition or laboratory analysis require authorisation in order to put in place the safety requirements posed by the legislation. In Slovenia the legislation regarding such materials is generally based on Euratom Treaty directives and no specific legislation regarding only radioactive geological samples is published. In addition, no international standards or guidelines exist according to the knowledge of authors in this area. Due to the fact that many private geological collections could have radioactive materials such guidelines would be beneficial.

2.3 Concentration of radioisotopes and authorisation of practice

The identified potentially radioactive samples shall be always further carefully investigated by the radiation expert using the model correlating the measured photon dose rate, shape and mass of a sample and concentration of radioisotopes. The concentrations of uranium and thorium in samples, expressed in units Bq/kg, are important data in order to identify the samples which are according to Slovenian legislation considered as radioactive material or radioactive waste.

Based on this data the owner of radioactive samples decides about future steps of a lifecycle of a sample. Three possibilities exist.

1. If an owner decides to use or just to store a specimen, the process of authorisation based on Slovenian legislation takes place.
2. When an owner decides that a radioactive specimen is a waste, the storage of such radioactive waste must be according to Slovenian legislation preformed by the Agency for Radwaste Management.
3. In case that the owner would like to sell or donate such a sample to already authorised private collector or institute the transaction should be authorised.

In practice no such transaction took place after the inspection of the SNSA in 2007.

3 CONCLUSIONS

In the extensive campaign of the SNSA in order to identify radioactive sources at research laboratories and institutes in Slovenia the inspection of geological laboratories, collections and storages took place in 2007. Many radioactive geological samples e.g. uraninite mineral specimen were identified. From an old storage of geological samples around altogether 100 kg of radioactive materials were transported to the Storage for radioactive waste. At the educational institution around 300 geological samples were identified as radioactive sources. Majority of them are still in use while a few hundreds of kilograms of radioactive waste originated from the educational institution were stored in the Storage for radioactive waste.

At the inspection it was regularly found that users did not know that samples are radioactive materials and no safety measures were in place. After the campaign the actual exposures of workers, students and visitors were efficiently lowered because the majority of sources were put in suitable storages and many safety precautions took place. In view of a lack of specific international standards or guidelines in this area, the authorisation of a practice related to radioactive geological samples shall be authorised as any other practice concerning radioactive materials or waste.
REFERENCES


