

RADIATION PROTECTION, REGULATORY AND WASTE DISPOSAL ASPECTS OF THE APPLICATION OF MINERAL INSULATION WOOL WITH ENHANCED NATURAL RADIOACTIVITY.

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1 ABSTRACT

Many scrap yards in The Netherlands have installed portal radiation monitors at the entrance for control on radiation sources in the scrap. If these monitors are triggered by a scrap load the material is refused and the appropriate Inspectorate is informed about the incident. Over the last few years these incidents have led to an increasing number of cases in which mineral insulation wool adhering to the scrap was identified as the radiation source. From these cases it appears that this wool most likely has been produced from a by-product (slag) of tin smelting between the years 1948 and 1960. It has been applied in widely ranging constructions and installations. Examples are insulation doors, bakery ovens, small steam generators and a large coal-fired power plant. Average activity concentrations in 40 samples of slag wool were 4 Bq/g and 11 Bq/g for nuclides from the U-238 and Th-232 decay chains respectively. The material was shown to be depleted in Pb-210 relative to Ra-226 and U-238. Under Dutch Nuclear Law waste with activity concentrations as indicated above has to be transferred to the Central Organisation for Radioactive waste at very high costs.

Radiation exposures at dismantling of slag wool insulated installations are estimated to remain below 2 mSv/a and are dominated by internal exposure by inhalation. Respiratory protection reduces the exposures to small doses from external exposure.

On the basis of the radionuclide concentrations and a survey on mineral wool production in the Netherlands there is strong evidence that the slag wool originates from slag of a tin smelter.

Tin refinement has taken place at a large scale in Western Europe in the past, in particular in Spain and the UK. In the latter country former sites of tin smelters with large inventories of tin slag have been identified and remediated. UK DETR estimates that 30 million ton of tin slag with enhanced levels of natural radioactivity has been applied in civil engineering in North West England.

With the implementation of the Council Directive 96/29 Euratom into Dutch Law the slag wool will still fall under the requirement for authorisation and when no regulatory action is undertaken the slag wool will still follow the golden route to COVRA. The very stable glassy character of the slag wool allows other disposal options such as landfills or repositories used for non-radioactive wastes. Apparently such radiologically and economically acceptable solutions can be found when the scale of the problem is large enough, as it is in the UK.

2 INTRODUCTION

Naturally Occurring Radioactive Materials (NORM) are encountered in many, sometimes surprising appearances. One way by which not seldom NORM containing materials are discovered is when scrap loads enter scrap yards or the premises of a smelter through a portal radiation monitoring system. These are installed mainly to protect against the processing of orphaned radiation sources which are a real economical and potentially a significant radiological risk. The alarm levels of these monitors can be set at dose rates above background as low as 5 – 20 nSv/h to optimise the chances that sources shielded to a large extent by the scrap load are still detected. Over the last few years a series of these alarms were caused by a mix of 'clean' scrap and insulation material with enhanced concentrations of natural radionuclides. From the beginning it was suspected that insulation material had been produced in the past with slag from some specific metal smelter. In all these cases the scrap was not accepted and usually returned to the owner and the incident was reported to the Inspectorate of the Environment. The incidents covered a variety of small to very large installations which had in common that once upon a time mineral wool had been used for thermal insulation of doors, tanks, ovens, and piping which now were dismantled. In most of these cases the presence of the mineral wool had serious economic consequences for the owners of the installations as a result of the application of the Dutch Nuclear Law. The accumulation of cases has led to a study to find out the origin and potential scope of what has since been called 'the slag wool problem'. Various aspects of this problem are discussed in this paper.

3 RADIOACTIVITY IN THE SLAG WOOL

A limited set of analytical data obtained by gamma spectrometry was available at NRG. A much larger set of data was made available by the Inspectorate for the Environment, from the reported incidents and their follow up. These data are summarized in Table 1.

Table 1. Natural radionuclides in slag wool samples from Dutch installations. Concentrations in Bq/g.

Type of data	U-238 and Ra- 226	Pb- 210	Th- 232nat	Total	ratio Th-232/U- 238	ratio Pb-210/Ra- 226
Number of results	40	32	40	40	40	32
Maximum	6.2	3.8	16	220	4.2	1.0
Minimum	2.6	1.7	5.9	40	1.8	0.4
Average	4.0	2.9	11.2	163	2.8	0.8
Stand. Dev.	0.8	0.5	2.1	28	0.5	0.1

The total concentrations in the table were calculated according to Dutch rules as $11 \cdot \text{U-238} + 3 \cdot \text{Pb-210} + 10 \cdot \text{Th-232}$. It appears that the concentrations of the radionuclides from the decay chains of both U-238 and Th-232 cover a relatively small range and that Pb-210 is depleted relative to U-238 and Ra-226. The average ratio Th-232/U-238 is about 3. From the average ratio Pb-210/Ra-

226 of 0.8 an average age of about 50 years can be derived if it is assumed that virtually all of the Pb-210 was volatilised at the time of the slag or slag wool production.

4 DUTCH NUCLEAR LAW AND SLAG WOOL

Under the, probably still current, Dutch regulations the rules for exemption and from the requirement of authorisation to possess and process materials with natural radionuclides are based on exemption total activity concentrations and total activities contained in one radiation source. The concentration limit is 500 Bq/g if the radionuclides can be assumed to be in their natural matrix. In other cases the same concentration limit of 100 Bq/g applies as for artificial radionuclides. The limit is applied to the total radionuclide concentration, including all short-lived radionuclides of the natural decay chains. The total activity limit depends on the radiotoxicity of the radionuclides involved. The longer-lived radionuclides from the decay chains of U-238(Th-234, U-234, Th-230, Ra-226, Pb-210 and Po-210) and Th-232 (Ra-228, Th-228) belong to the high radiotoxicity group for which an exemption level of 5 kBq/g applies. Raw materials and residues from the non-nuclear industry usually exceed the total activity limit because of the amounts of material involved. The concentration limit is the deciding one with respect to the requirement for authorisation.

Wastes from the non-nuclear industry which exceeds the activity concentration limit may only be disposed of by transfer to the only organisation authorized to receive radioactive waste: the Central Organisation for Radioactive Waste (COVRA).

Using the criterion of 'radioactive equilibrium in a decay chain is natural' and taking into account the depletion of Pb-210, and consequently also Po-210, in the slag wool, the total activity limit of 100 Bq/g applies. From the ranges and the average of the total activities in slag wool presented in Table 1 it is clear that most of the slag wool detected in the 40 incidents exceeded the 100 Bq/g level. Consequently, the surprised owners of the material formally are required to apply for an authorisation and COVRA is the only legal route for disposal.

5 CONSEQUENCES OF CURRENT DUTCH LAW

Although the competent Dutch authorities do not stick to the formal authorisation requirement, they do require the removal of the slag wool from already disused objects or from the still remaining parts of the installations being dismantled. The removal has to be supervised by qualified radiation protection experts under precautions against internal exposure not unlike those taken at removal of asbestos.

The slag wool incidents cover steam generators for greenhouse heating and small industrial applications, a bakery oven, an oven at a foundry, insulating doors at a steel and a roof tile factory and a huge power plant. Three of these case are chosen to illustrate the Dutch slag wool problem. They have in common that the slag wool was detected, as usual, by a scrap dealers' radiation detection system and that the costs for transfer of the slag wool to COVRA are based on a realistic estimate of about 18 Euro per kg (40 NLG per kg)..

- Greenhouse steam generator

The slag wool from this relatively small steam generator was suspected of exceeding the 100 Bq/g level on the basis of readings with hand-held radiation monitoring equipment. The estimated COVRA costs were of the order of 18,000 Euro, exclusive of the costs for removal of the slag wool under radiological control and packing it into the standard 100 liter COVRA drums. Upon analysis of a slag wool sample by gammaspectrometry the total activity concentration appeared to be just below 100 Bq/g and the steam generator was exempted from radiological control.

- Bakery oven

Upon detection of radioactive slag wool during dismantling of a bakery the already advanced dismantling was continued under radiological control and the total amount of scrap mixed with slag wool was transported from the site for removal of the slag wool and packing into COVRA drums. Eventually a little short of 100 COVRA drums resulted for which the costs were estimated at about 105,000 Euro, exclusive of the significant costs of getting the slag wool separated from the scrap and into the COVRA drums. The drums still rest at NRG until the financial issues are solved.

- Power plant

Slag wool had also been used for insulation of piping and huge boilers in a big fossil fuelled power plant. It took many month first to collect slag wool from radiologically uncontrolled dismantling and to finish the dismantling under radiological control. In total 36 tons of slag wool compressed and packed in plastic bags resulted from the exercise. They are still stored at the site. The costs of transfer the slag wool to COVRA after overpacking into drums are estimated at about 650,000 Euro.

6 RADIOLOGICAL ASPECTS OF DISMANTLING SLAG WOOL INSULATED INSTALLATIONS

The presence of slag wool in installations being dismantled involves external exposure as well as internal exposure by inhalation of slag wool particles. External dose rates on the surface of slag wool insulated equipment does not exceed 1 $\mu\text{Sv/h}$ and an average dose rate at working distance of 0.3 $\mu\text{Sv/h}$ seems to be a good estimate for dose assessments. The radiation doses resulting from internal exposure depends strongly on whether the radioactive character of the slag wool has been recognised before dismantling was started. As is shown in Table 2 the dose coefficient for inhalation of slag wool particles with average and high nuclide concentrations ranges between 1 and 1.5 mSv per gram inhaled. The main contribution to internal exposure comes from the Th-232 decay chain.

On the basis of the basic data for internal and external exposure described above dose estimates can be made for three different exposure scenarios defined in Table 3. It is assumed that slag wool removed from the installation is either temporarily stored remote from the working place or is disposed off frequently.

Tabel 2. Dosiscoefficienten (DC) for inhalation of nuclides from slag wool based on 5 µm AMAD and Type Slow.

Nuclide	DC in Sv/Bq	Average conc. Bq/g	DC in Sv/g inhalation	High conc. Bq/g	DC in Sv/g inhalation
U-238 - Ra-226	$6.4 \cdot 10^{-5}$	4	$2.6 \cdot 10^{-4}$	6	$3.8 \cdot 10^{-4}$
Pb-210 - Po-210	$8.1 \cdot 10^{-6}$	3	$2.4 \cdot 10^{-5}$	4	$3.2 \cdot 10^{-5}$
Th-232 - Po-212	$6.3 \cdot 10^{-5}$	11	$6.9 \cdot 10^{-4}$	16	$1.0 \cdot 10^{-3}$
Totaal			$9.7 \cdot 10^{-4}$		$1.4 \cdot 10^{-3}$

Table 3. Scenario conditions and parameters for external and internal exposure to slag wool at dismantling of installations.

Scenario	Radio-activity recognised	Size of installation	Exposure hrs/a	Slagwool dust conc. mg/m ³	Breathing rate m ³ /h
A	no	small	20	1	1.2
B	yes	big	200	n.r. ¹⁾	n.r. ¹⁾
C	no	big	400	2	1.2

¹⁾ Not relevant because of assumed respiratory protection

The results of the scenario calculations are presented in Table 4. They show that under unfavourable conditions but average activity concentrations the radiation exposure of demolition workers is estimated at about 1 mSv/a. For the maximum concentrations given in Table 1 the estimated exposure does not exceed 2 mSv/a.

Table 4 Estimated radiation exposures to slag wool at dismantling of installations.

Scenario	External exposure mSv/a	Internal exposure mSv/a	Total exposure mSv/a
A	0.006	0.024	0.03
B	0.06	-----	0.06
C	0.12	1.0	1.1

7 ORIGIN OF THE SLAG WOOL

KEMA, in cooperation with NRG, has carried out a survey on the origin and potential extend of the slag wool problem under contract with the Dutch Government. In an early stage of the survey it appeared that the radionuclide concentrations and the ratio between radionuclides of the U-238 and Th-232 decay series matched well with slag from a Dutch tin smelter stored at COVRA after shut down of the smelter. Further inquiries revealed that producers of mineral insulation wool indeed have used slag from the Dutch tin smelter during a limited period of time, probably between 1946 and 1960. The bulk of mineral wool produced over those years was made from other raw materials, slag from

steel production and basalt with much lower concentrations of natural radionuclides. The KEMA survey estimates the total amount of mineral wool produced with slag from tin smelting at between 800 and 2200 tons.

8 INTERNATIONAL ASPECTS

Up to now no written evidence exists that the use of mineral wool from tin slag has drawn attention internationally for the same reasons as in The Netherlands. Tin production has taken place on a large scale in the past in Western Europe, particularly in the United Kingdom and Spain [2]. Most of the ore originates from South East Asia where low grade tin ore is processed by physical methods to obtain a concentrate (cassiterite) and a by product (amang). Tin ore not only contains the tin bearing cassiterite but also other heavy minerals including ilmenite ($\text{FeO}\cdot\text{TiO}_2$), zircon (ZrSiSO_4) and monizite ($[\text{Ce}, \text{La}, \text{Y}, \text{Th}]\text{PO}_4$) which all contain the naturally occurring radioactive elements thorium and uranium [3]. The potential for significant radiation exposure of workers and the public in the tin mining, ore preparation industry and tin smelter in Malaysia has been recognised [3], [4], [5].

A study by Baxter et al. summarises unpublished reports on a large tin smelter in Northern England [6]. Although the report focussed mainly on discharges of and exposures to Po-210 volatilised from the raw material, it contains interesting information on the scale of the slag production at the site. Average annual intake of raw materials was 82,000 tons and the average annual output of waste slag was 60,000 tons. The slag from a storage tip on the site was sold periodically for road construction or cover for a domestic refuse site. Some was deposited in a local landfill site and some sold to a local abrasive manufacturer. Cleaning up the site at demolition involved disposal of demolition debris and waste slag in a landfill site or the local authority waste disposal site.

At another former site of a tin smelter in the UK in Bootle near Liverpool radiation levels over 10 times normal background were detected on rocks in the river estuary [7]. The rocks appeared to be made up of waste slag from the local smelting works. It is interesting to note that the critical group of members of the public were likely to be dog owners exercising their animals in the beach area. Because of development plans for the site of the former tin smelter a survey was carried out which revealed dose rates up to $10 \mu\text{Sv/h}$ at 1m above the surface. It was found that the area was covered with slag and rubble to depth of about 1.5m. About 1000 m^2 was covered with about 1900 tons of a more active black slag. The concentration of U-238 in four samples of this black slag ranged from 5.0 to 6.2 Bq/g and the Th-232 concentration ranged from 12.1 to 14.7 Bq/g. The average Th-232/U-238 ratio was 2.4 ± 0.2 . The total activities were estimated at 11.3 GBq U-238 and 26.9 GBq Th-232. The authors advised to fence off the area and clear the area under radiological control of the workers and to find a disposal route for the excavated slag and rubble.

In a Radioactivity Information bulletin the UK DETR provides an estimate of 30 million tonnes of glass like tin slag has been used as aggregate for general civil engineering in North West England and adds that the potential for harm from this material is likely to be low [8].

9 DISCUSSION AND CONCLUSIONS

Demolition of slag wool insulated installations are not likely to have caused excessive radiation exposures even when the radioactive character of the material was still unnoticed. However, respiratory protection should be a standard measure against avoidable exposure during demolition. The main aspect of the slag wool problem in the Netherlands is the excessive cost of its transfer to COVRA for long-term interim storage and still undefined final disposal. With the implementation of the Council Directive 96/29 Euratom into Dutch Law the slag wool will still fall under the requirement for authorisation and when no regulatory action is undertaken the slag wool route will still be to COVRA. It would be difficult to justify these costs to individuals and society on the basis of the radiation exposures being avoided by this disposal route. The very stable glassy character of the slag wool allows other disposal options such as landfills or repositories used for non-radioactive wastes. Apparently such radiologically and economically acceptable solutions can be found when the scale of the problem is large enough, as it is in the UK.

10 REFERENCES

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