Disposal and re-use of TENORM - legal limitations and obstacles

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Abstract:

While implementing EURATOM guideline 96/29 in the German legislation, in June of 2001 an essential pre-condition was created for re-use or disposal of TENORM. An essential progress has been achieved allowing to re-use TENORM or to dump it together with other residues and waste, if the specific activity does not exceed the limits defined in the Radiation Protection Ordinance (StrlSchV). Otherwise, if the specified limits in terms of concentration or radiation dose are exceeded, than these materials must remain under radiological protection.
A practical application of the new German regulation turns out to be difficult especially for disposal together with other waste and for re-use as backfilling material in mines taking into account problems arising from adaptation of the respective legislation on radiation protection, soil protection, waste management and shipment of dangerous goods.
The report tackles obstacles for re-use and disposal of TENORM together with garbage and toxic waste arising from the new legislation. Otherwise, proposals will be given how obstacles of selected options for re-use and disposal can be overcome.

1 INTRODUCTION

The general provisions as well as the single paragraphs of the German Radiation Protection Ordinance (StrlSchV) which are directed towards natural radiation sources are based on recommendations of international organisations, especially according to Title VII of the Directive 96/29 EURATOM, but also according to radiological protection principles developed by the German Commission on Radiological Protection (SSK). Accordingly, radiation protection measures must be taken into account if the existence of natural radiation sourcers could lead to enhanced radiation exposure. For that purpose regulations were agreed. According to the StrlSchV materials which contain natural radionuclides are divided into residues (§97) and other materials (§ 102). Residues are defined as materials which are resulting from different technological processes listed in Annex XII part A of StrlSchV with specific activity of at least 0.2 Bq/g. This list comprises sludge and scales from the exploitation of natural gas and crude oil and residues and by-products from processing of raw phosphate, iron and non-iron ores and minerals. Other materials are considered as radioactive material only if they are a source of enhanced exposure and countermeasures must be taken into account by the responsible Authority.

The defined concentration level for “enhanced” is derived from the annual dose limit for members of the public considering basic exposure scenarios. In Germany and some other EU member states this dose limit is 1 mSv/yr from which a minimum specific activity of 0.2 Bq/g per radionuclide of the U 238 decay chain and the Th 232 decay chain was derived.

Subsequently, natural or processed materials with lower specific activities turn out from radiation protection regulation.

The release of residues from radiological supervision for re-use or disposal according to § 98 StrlSchV or of other materials according to § 102 StrlSchV is connected with regulations of other legal grounds. That concerns provisions of the Waste Management Act (KrW-/AbfG) including the Ordinance on Utilization of Waste for Landfill Construction as well as regulations based upon the German Mining Act, e.g the Ordinances for Backfilling of Mines (VersatzV). The revision of the VersatzV in 2003 was connected with the reduction of the maximum permissible concentrations of some pollutants. Consequently, this option for re-use of radioactively contaminated residues became strongly limited because many of the concerning TENORM contain heavy metals, arsenic and sometimes organic compounds.

2 AMOUNT AND ACTIVITY LEVELS OF TENORM

The report [1] contains an overview on the kind, amount and range of specific activities of TENORM in Germany according to Annex XII StrlSchV. The following explanations are directed towards TENORM according to Annex XII with priority in terms of amount and activity level as well as of other materials according to § 102 StrlSchV of any relevance.

2.1 Scales and sludge from oil and gas extraction

Naturally occurring radionuclides are present in varying concentrations througout hydrocarbon reservoirs. The type of rock formation largely determines the radionuclide content of the reservoir; sedimentary rocks show low radionuclide concentrations whereas phosphate rock types have high levels. The waters associated with the rock formations often contain barium, strontium and radium. During the oil and gas extraction barium precipitates as sulfate inside the tube. Radium isotpes are co-precipitating due to their chemical similarity to barium. Consequently, the radioactivity of the scales result primarely from radium isotopes.

Based on a roughly calculation in [2] the annual amount of scrap from all German on-shore oil and gas extraction units is about 10.000 t containing approximately 1.000 t scales. The rise of sludge containing more than 100 Bq/g cross activity is given in [3] with 400 m^3/yr per off-shore oil extraction unit and 50 m^3/yr per on-shore extraction unit. The amounts of contaminated sludge from natural gas extraction are ten times lower.

2.2 Residues from bauxite porcessing

The processing of bauxite to aluminium oxide by means of the Bayer method lead to the by-product „red mud“ which consists mainly of iron oxide and silicates. The only alumina plant in Germany (“Aluminium Oxide Stade”) is processing about 1.5 millions tons of bauxite per year to produce 800,000 t aluminium oxide and 630.000 t red mud per year. The total amount of red mud is dumped at a landfill at the production site.

Bauxite is imported from China, Guinea, Guayana and Australia. The specific activity of red mud is between 0.05–0.7 Bq/g of U238, 0.02–1.6 Bq/g of Ra 226 and 0.05–1.0 Bq/g of Th 232 in dependance of the origin of bauxite.

2.3 Zirconium sands

Zirconium sands are used as foundry sand, for the production of refractory bricks, ceramics, glazes and enamels as well as optical glasses.

For milled zirconium sand, the U 238 activity is between 2.5 and 4.0 Bq/g and the Th 232 activity between 0.6 and 0.7 Bq/g. The specific activity of refractory bricks can rise up to 10 Bq/g of U 238 whereas ceramic products show U 238 activities up to 6.0 Bq/g.

According to [3] the amount of spent foundry sands (zirconium sand) is about 5,000 t/yr and of firebricks from furnace, scouring and blasting sands as well as smelting and foundry debris about 1.3 millions t/yr. Their mean specific activity is in most case lower than 0.5 Bq/g, because the bigest part of these materials have no enhanced level of natural radioactivity.Therefore these residues are commonly delivered to public landfills.

2.4 Dust from primary metal smelting

During the processing of iron ores and non-iron ores some radionuclides are enriched in filter dust. That concerns the volatile radionuclides Pb 210 und Po 210. According to [1] the maximum specific activity of 8.5 Bq/g Pb 210 was measured in dust samples from iron blast furness.

2.5 Other materials

Other materials according to § 102 StrlSchV with higher quantities and specific activities of more than 1 Bq/g are sludge from water treatment and filter dust from secondary smelting processes. Furthermore, the discharge of pit water from hard coal mines lead sometimes to contamination of river bank sediments. According to [3] a maximum specific activity of 30 Bq/g Ra 226 was measured in flood plain soils.

3 RECENT PRACTICES FOR RE-USE AND DISPOSAL

3.1 Re-use or disposal of residues from oil and gas extraction

Since a couple of years different methods for re-use and disposal of sludge and scales from gas and oil extraction are applied or the licensing process is running. The preference of an option depends e.g. from the radioactivity level and the presence or absence of mercury. The following options for re-use or disposal of contaminated scrap, of separated scales and of sludge are applied.

Untreated tubes could left in place, but not feed pipes due to restrictions by the German Mining Act.
In the past, tts dumping on common land fills, clay pits or holes from open pit mining was only applicable for tubes with a specific activity of the scales of lower than 0.2 Bq/g and if mercury is absent. Nevertheless, the smelting of the tubes was preferred due to economical reasons.

Recently, also higher contaminated scrap could be dumped on landfills according to Annex XII part C of the StrlSchV if mercury is absent. The dumping at land fills of low contaminated scrap with a total activity of the scales between 0.2 Bq/g and about 1 Bq/g was allready executed. But the portion of such tubes is to small to apply this option on a broader scale. Nowadays a licensing process is running for dumping of scrap containing both, radionuclides with higher specific activities and mercury at a special landfill for toxic waste.

During the last three years GRS worked out an option for the disposal of scrap poluted by both, natural radioactivity and mercury in an underground disposal site for toxic waste [4] basing on the dose criteria according to Annex XII part D of StrlSchV in connection with the regulations for toxic waste management and the shipment of dangerous goods. This option is applicable for a cross activity of the scales up to 40 Bq/g. Now the licensing process is interrupted by the Mining Authority of Saxony-Anhalt due to intervention by the owner of this underground disposal site which is a separate part of an operating potash mine. Therefore huge amounts of scrap contaminated by both, radioactivity and mercury are recently piled up at storage areas of the oil and gas companies.

At the moment, the smelting of higher contaminated scrab can only be carried out by the Siempelkamp company having a licence for smelting of such scrap. This option is limited due to very high costs.

The scales can be separated from the tubes by means of high pressure water jet technique. The application of this technique becomes increasing importance. The remaining radioactivity of the cleand up tubes is lower than 0.1 Bq/g. The unrestricted re-use of such tubes should be possible but at the moment only its smelting is licensed by the responsible State authorities.

For the re-use or disposal of the separated scales different methods are applied. The common method was the backfilling of the scales into the boreholes. Recently this method was stopped by the responsible Mining Authority due to stronger demands for the long term safety, i.e. for the exclusion of the migration of mercury into the groundwater. Nevertheless, some geological formations where oil and gas extraction takes place fulfil these conditions and backfilling takes place again.

Sludge from oil and gas exploitation is commonly dumped on special landfills. Since a couple of years a method for demercurisation of sludge from oil and gas industry is applied. Recently this method was modified for scales. According to [3] the recycled mercury is of highest purity. After the selectiv purification the sludge or the scales still contain the same specific activity as before. These residues - in case of sludge after partial dewatering – are mixed with a Geopolymer^{R} (high-alkali (K-Ca)-Poly(sialate-siloxo) binders) for immobilisation. The sludge or the scales, that means waste, are going to be again a waste after demercurisation, but on the one hand an important raw material (mercury) is extracted and on the other hand a high toxic substance is together with hydrocarbons removed. The radio-nuclides are safely encased for the long-term due to conditioning by means of Geopolymer^{R} . The storage of the immobilized residues at landfills or underground dump sites is possible in accordance with the legal frame conditions.

3.2 Re-use or disposal of residues from bauxite processing

The disposal of red mud at a landfill on the AOS site will be continued. The remaining capacity of this landfill is big enough for storaging the yearly amount of red mud for some years.

3.3 Re-use or disposal of spent zirconium sand and residues

As already explained the bigest part of spent foundry sands and fire bricks have no enhanced level of natural radioactivity, because zirconium sands represent only a minor part of these materials. Therefore such residues are commonly delivered to public landfills. Nevertheless, the existance of spent residues made of zirconium sand with enhanced levels of natural radioactivity can not be excluded. The knowledge on the amount of such materials is poor due to highly complicated reconstruction of the pathes from zirconium mills to appearance of spent material.

3.4 Re-use or disposal of other materials

Other materials, as filter dusts, ashes and slags from ore porcessing are commonly dumped together with other waste on landfills for garbage or demolished building materials.

4 LEGAL FRAME CONDITIONS FOR RE-USE OR DISPOSAL OF TENORM

The above described methods for re-use or disposal of some TENORM are only applicable if they are in agreement with the legal frame conditions. That concerns not only the regulations on radiological protection but also those on recycling and storaging of common residues and waste as well as on the shipment of dangerous goods. In every case – except for the controlled smelting by Siempelkamp Co. – the precondition for the implementation of each practice is the release of the concerning residue from the radiological supervision according to § 98 of StrlSchV in connection with Annex XII, part A (kind of material) part B (maximum permissible specific activity), part C (maximum permissible specific activity of the total material of an landfill) and/or part D (keeping of the dose limit of 1 mSv/yr).

The procedure necessary for the release of such materials from radiological supervision is much more complicated than assumed, because the regulations for the different regulations are not well tuned or some regulations changed since the StrlSchV was set in force.

4.1 Joint disposal with common waste

The Radiologial Protecton Autority can release such residues from supervision if the above mentioned conditions are fulfiled. On the other hand, in case of joint disposal with common waste the Autority which is responsible for common waste affairs must allocate a single disposal facility. The owner of this facility can only accept such material after its release from radiological supervision. For that reason the most of licencing processes are not yet finally decided, especially if Authorities from different German states are involved into this process or if materials come from abroad.

According to the Ordinance on Waste Management (KrW-/AbfG) the re-use have priority against disposal. Disposal options are only allowed if no option for re-use exist. Consequently, the amount of waste for disposal is decreasing with the aim to finis the waste disposal in 2020. According to Annex XII part C of StrlSchV the precondition for joint disposal of residues with common waste is given if the mean specific activity of these radionuclides of the uranium-radium decay chain and the thorium decay chain with the highest value of activity devided by the total amount of waste dumped per year on the concerning disposal site is lower or equal to the defined limits. Consequently, as less conventional waste is dumped per year also less amount of radioactively contaminated residues can be disposed on landfills. Considering the termination of this disposal option more and more landfills will be closed.

The option of underground disposal of toxic waste will be continued. Nevertheless, the amount of such waste is lower than previously assumed. Furthermore, in case of disposal of toxic waste in underground mines mixing of different kind of waste is prohibited to avoid chemical reactions between components of these waste, i.e. the formula for mixing given in Annex XII part C of StrlSchV is not applicable. For that reason the proof for this disposal option must be made via dose calculation to fulfil the dose criterion of 1 mSv/yr.

4.2 Re-use

Building materials

The re-use of residues listed in Annex XII part A of StrlSchV as building material is limited in Germany because the concerning recommentation of EC [5] was not recommended for national application. It can be expected that this option for re-use will be finished totaly if the draft of the Ordinance on Indoor Radon Protection will come in forces.

Only the re-use of copper slag was applied. Allready in 1991 the German Commission on Radiological Protection (SSK) recommended the re-use of copper slag for the construction of roadbeds [6]. Nevertheless, the company which processes this material needs a licence from the Radiation Protection Authority based upon the Radiation Protection Ordinance (VOAS) of the former GDR. Furthermore, for the re-use of this material this authority had to grant a permission on the basis of the Order for re-use of contaminated materials (HaldAO). Both regulations remained their validity till 2001 on the territory of the former GDR. Since the implementation of the new Radiation Protection Ordinance (StrlSchV) in 2001 the permission for the re-use of the copper slag was redrawn by the State Authority of Saxony-Anhalt, although it would be possible if considering only the limit of the radionuclide concentration given in this new regulation.

Backfilling material, above ground

In that context the Ordinance on Re-use of Waste as Construction Material for Landfills [7] and the rules for the use of waste for covering of waste rock heaps [8] must be considered.

According to this Ordinance wastes and residues can be applied as substitute of material for the construction and recultivation of landfills as far as they have specific mechanical properties and do not exceed maximum permissible concentrations of some pollutants (s. Table 1). Althought many of the residues listed in Annex XII part A of StrlSchV especially such arising from mining and processing of metal ores show the necesasary physical properties this option is limited due to accompanying high concentrations of heavy metals or arsenic. That is the same for the re-use of such materials for covering of waste rock heaps and tailings ponds from salt and coal mining (s. Table 1). Only foundry sands including zirconium sands can be commonly used as cover material according to [8].

Table 1 Maximum permissible concentrations of selected parameters according to [7], [8]

Parameter	Maximum concentration [mg/kg]		
	1	2	3
Arsenic	20	30	50
Lead	100	200	300
Cadmium	0,6	1	3
Copper	40	100	200
Mercury	0,3	1	3
Sulfate	50	50	100

1 = for landfill basement [7] or unrestricted re-use (re-use class W 0 [8])
2 = as mineral sealing layer [7] or restricted open re-use and consideration of groudwater protection (re-use class W 1.1 [8])
3 = as pure mineral sealing layer [7] or restricted open re-use (re-use class W 1.2 [8])

The values given in Table 1 are in compliance with the German Soil Protection Act for comparable scanarios.

Backfilling material, underground

The backfilling of boreholes from oil and gas exploitation ist limited due to the new requirements for the long-term safety.

As for the above ground use of wastes also for the re-use as backfilling materials in underground mines the maximum permissible concentration for some pollutants have been reduced in the new Ordinance on Backfilling of Underground Mines [9]. Furthermore, the preferrence of metal extraction from waste against the use as backfilling material is given if defined metal concentrations are exceeded (s. Table 2).

Table 2 Limits for metals in waste (based on dry weight) and limits for solids as well as concentration in eluate according to [9]

Parameter	Limits in waste		
	Metals [g/kg]	Solids [mg/kg]	Eluate [µg/l]
Zinc	≥ 100	1.500	500
Lead	≥ 100	1.000	25
Copper	≥ 10	600	50
Tin	≥ 15	-	-
Chrome	≥ 150	600	50
Nickel	≥ 25	600	50
Iron	≥ 500	-	-
Cadmium	-	10	5
Mercury	-	10	1
Arsenic	-	150	10

4.3 Shipment of dangerous goods on roads

A further problem is rising up from the regulations for shipment of dangerous goods. In Germany it is not allowed to store such radioactive waste and contaminated residues (TENORM) at disposal facility for toxic waste which are transported under the regulation for shipment of radiactive materials on roads (ADR). This option for disposal is interupted because the maximum permissible concentration of natural radinuclides for release from radiological supervision may be higher than the minimum specific activity according to the regulation on transportation of radiactive materials. Whereas according to the StrlSchV the release is based on the maximum specific activity of that nuclide with the higest activity, for transport issues the sum of all long living radionuclides must be considered. For radioactive equilibrium this value is 100 Bq/g but for non-equilibrium conditions it depends from the portion of radioactive thorium isotopes having a 10times lower exemption limit than e.g. radium or lead isotopes. That results in a “flowing minimum permissible activity” for shipment as the following figure demonstrates.

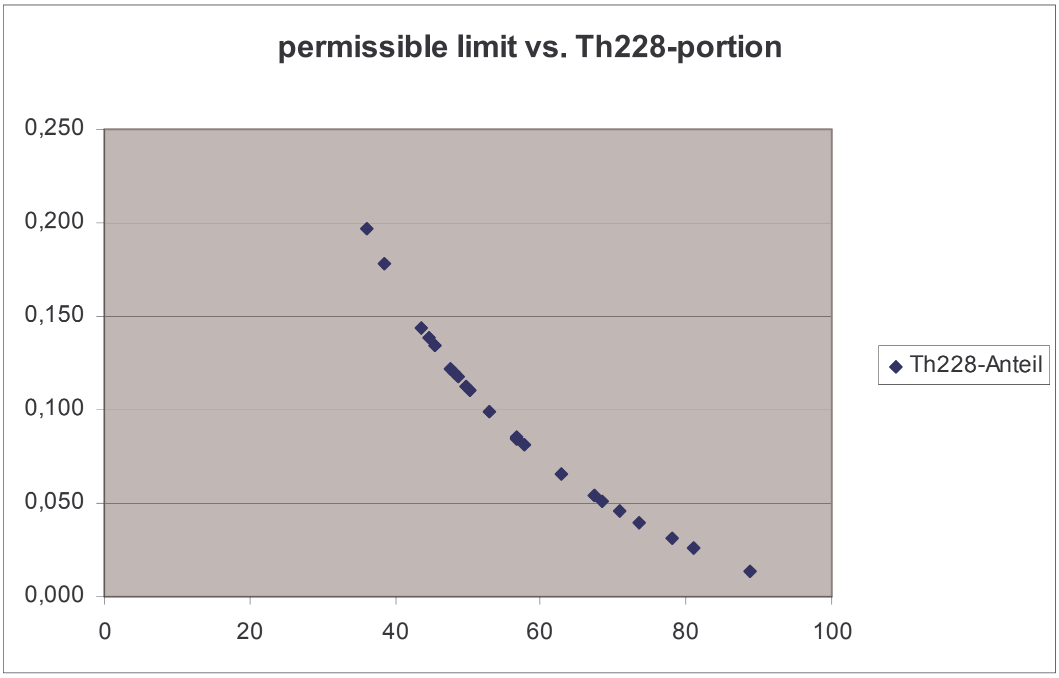

Figure 1: Minimum cross activity for release from transportation regulation in dependance of the portion of Th 228 against the portion of Ra 226, Ra 228, Pb 210 and Po 210 [8]

In case of scales from oil and gas exploitation the contribution of thorium isotopes to the gross activity depends from the hydrogeological conditions of the deposit but it is relatively cnxstant at one deposit. Consequently, the real limit of gross activity for exclusion from shipment according to ADR class 7c depends from the origin of the tubes. Whithout doubt, a "variable limit" is a challenge for the licensing body.

5 SUMMARY AND CONCLUSIONS

- The most of TENORM can be released from radilogical supervision according to §§ 98 and 102 StrlSchV in connection with Annex XII, part B, C and D.

- The main problems for the licensing process arising from the superposition of different legal aspects which have to be taken into account.

- There is not only one preferred option for re-use or disposal of each kind of TENORM, but it depends from different factors as
  - level of the specific activity or
  - presence and concentration level of chemical toxic substances

- The expected reduction of landfill capacity, the stronger limits of pollutant concentrations for re-use of residues as back-filling material and the limitation for re-use as building material according to some new or revised regulations will lead to a loss of some radiological "safe" options for re-use as well as for disposal.

- Nevertheless, the annual amounts of TENORM are lower than expected and the overall storage capacity of landfills is still sufficient.

- If applying immiobilisation measures the capacity of suited storage grounds could be enlarged due to reduction of radionuclide leaching from immobilised residues.

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