TREATMENT AND DISPOSAL OF NORM AT SPECIAL LANDFILL SITES AND FORMER URANIUM MINING SITES IN GERMANY: PRACTICAL APPROACHES AND SOLUTIONS

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1 ABSTRACT

Due to their volumes and specific activities, for a part of NORM (Naturally Occurring Radioactive Materials) a release from regulatory control is not possible without further measures. Instead, their treatment and disposal require the immobilisation of the radionuclides as well as special technologies for their disposal. For the disposal of large quantities of residues with elevated radioactivity resulting from the treatment of contaminated water at former uranium production sites in East Germany, the companies B.P.S. Engineering GmbH and WISMUT GmbH have developed suitable technologies. The immobilisation of residues with specific activities up to several hundred Bq/g of the main nuclide can be achieved by GEOPOLYMER®, an inorganic hydraulic binder. Disposal sites were designed and constructed which satisfy radiological requirements and requirements in accordance with water protection regulations. The paper presents these technologies and disposal sites and shows their applicability to treatment and disposal of other NORM.

2 INTRODUCTION

German radiation protection legislation has recently been brought in line with the EU Directive 96(29 (1). The amended Radiation Protection Ordinance brings new regulations for the protection of the population with respect to natural radioactivity. In particular, § 97-99 focus on the handling of residues contaminated with natural radionuclides. The treatment and disposal of NORM fall into this category. These residues can be released from regulatory control if the exposure of individuals does not exceed 1 mSv (a after disposal or re-use of the residues. For a part of materials due to their elevated specific activity immobilisation techniques must be applied, and disposal has to be facilitated in such a way that the 1 mSv requirement is satisfied.

3 REGULATIONS ON RELEASE OF RESIDUES CONTAMINATED BY NATURAL RADIONUCLIDES FROM CONTROL

Annex XII to the new German Radiation Protection Ordinance defines a stepwise procedure for the evaluation of residues with elevated natural radioactivity with regard to their possible release from regulatory control. This procedure is based on the following test criteria (1):

Step 1: For decision whether the material has to be considered as a “residue to be subject to inspection” or not: Assessment of the residues with regard to their origin (list of residues according Annex XII A of the new Radiation Protection Ordinance) and to their specific activity (has to exceed 0.2 Bq(g for at least one nuclide of the U-238 or Th-232 decay chain)

Step 2: Inspection limit for residues to be considered: For re-use or disposal the sum of the highest specific activities $C_{U238max}$ and $C_{Th\text{-}232max}$ of the nuclides U-238sec and Th-232sec of both decay series must be lower than a limit C (cf. Annex XII B of the Ordinance):

$(R \cdot) C_{U238max} + C_{Th232max} \leq C$ with:
C = 1 Bq(g
C = 0,5 Bq(g for disposal of > 5 kt(a in feeding areas of usable aquifers
C = 5 Bq(g for underground disposal
($R \neq 1$ only for elevated $C_{Pb210++}$)

Step 3: Release from regulatory control in case of co-disposal of radioactive residues with other waste: The sum of the respective averages $C^M{}_{U238max}$ and $C^M{}_{Th\text{-}232max}$ of the total of residues must not exceed a limit C^M (cf. Annex XII C of the new Ordinance)

$(R \cdot) C^M{}_{U238max} + C^M{}_{Th232max} \leq C^M$, with:
C^M = 0.05 Bq(g, for disposal at landfills > 15 ha
C^M = 0.1 Bq(g, for disposal at landfills ≤ 15 ha
C^M = 5 Bq(g, for underground disposal (R see above)

If a substance is subject to inspection following step 1 and if criteria according to step 2 or step 3, respectively, are satisfied it may then be assumed that the disposal without additional measures will not entail radiation exposure in excess of 1 mSv(a for a member of the population at large. In such case, the residues (NORM) might even be placed in a conventional landfill (1).

It may be anticipated that the practical implementation of this procedure will bring up a number of questions, e.g. regarding the determination of the averages $C^M{}_{U238max}$ and $C^M{}_{Th\text{-}232max}$. Problems will arise in particular when the limits C or C^M will be approached or even exceeded. An analysis reveals that this will be true for a considerable part of NORM produced (2,3). In such a case, the person in charge of residue handling would have to take action and furnish proof that the intended disposal will also not result in doses in excess of 1 mSv This results, as a rule, in the need to immobilise radioactivity and in more stringent requirements with regard to the disposal site. In the following, feasible approaches are presented for such cases, including the handling of residues from the treatment of contaminated waters at WISMUT.

4 RADIOACTIVELY CONTAMINATED RESIDUES FROM WATER TREATMENT OPERATIONS AT WISMUT

Since 1991 WISMUT GmbH is conducting the remediation of the legacy of more than 40 years of intense uranium mining in East Germany. Treatment of contaminated mine waters as well as of seepage and pore waters from mine dumps and tailings ponds constitutes one of the major challenges facing WISMUT in their efforts to gradually reduce environmental impacts. Water treatment generates considerable volumes of residues with elevated specific activities (cf. Table 1). These residues have to be safely deposited in full compliance with radiation and water protection requirements.

Table 1: Residues from water treatment at WISMUT GmbH

Site	Current waste generation per year [t]	Plant operation anticipated to end in: [year]	Specific activity (order of magnitude)	
			U-238 [Bq(g]	Ra-226 [Bq(g]
Aue	3.200	2030	500	20
Pöhla	50	2010	< 1	40
Helmsdorf	2.900	2025	100	< 5
Königstein	15.000	2012	60	10

(Start-up of new water treatment plants at the sites Seelingstädt and Ronneburg is planned for the second half of year 2001, respectively for the first quarter of 2002.)

The companies B.P.S. Engineering and WISMUT have developed technologies for disposal of the residues from WISMUT water treatment plants. The technologies were also applied to the disposal of residues from areas outside of the WISMUT remediation project, e. g. to residues from the reprocessing (demercurization) of scales from oil and gas pipes. Immobilisation of residues with specific activities from natural radionuclides of up to several hundred Bq/g of the main nuclide can be achieved by using GEOPOLYMER®, an inorganic hydraulic binder (4,5). This type of immobilisation (GEOPOLYTEC®-Technology) is explained in more detail in the following section.

5 GEOPOLYTEC® TECHNOLOGY

For sludges from water treatment facilities operated by WISMUT as part of its mining remediation activities and for residues from other industrial processes such as demercurization of scales from oil and gas pipes, regulators require radionuclides and other hazardous components to be immobilized with strong emphasis on long-term stability. Requirements for the design of the disposal site are closely linked to the stability of the waste packages stored therein. In search of a solidification method that would both be cost-effective and satisfy regulatory requirements, high-alkali (K-Ca)-Poly(sialate-siloxo) binders which have been coined Geopolymers have been identified as a viable solution. GEOPOLYMER®, despite its perhaps misleading name, is a purely inorganic material resulting from a polycondensation reaction ("geopolymerisation") yielding three dimensional zeolithic lattices. The terminology pertinent to the chemistry of Geopolymers which have been rediscovered by Davidovits and co-workers in the 80's can by found in (4). The basic chemical structure of Geopolymer is shown and compared to that of Portland cement in Figure 1. The patented solidification technology on the basis of GEOPOLYMER® has been coined GEOPOLYTEC® process. Other authors have previously investigated the use of Geopolymers to solidify toxic wastes (6(before the first results on the solidification of NORM wastes were published in (7(.

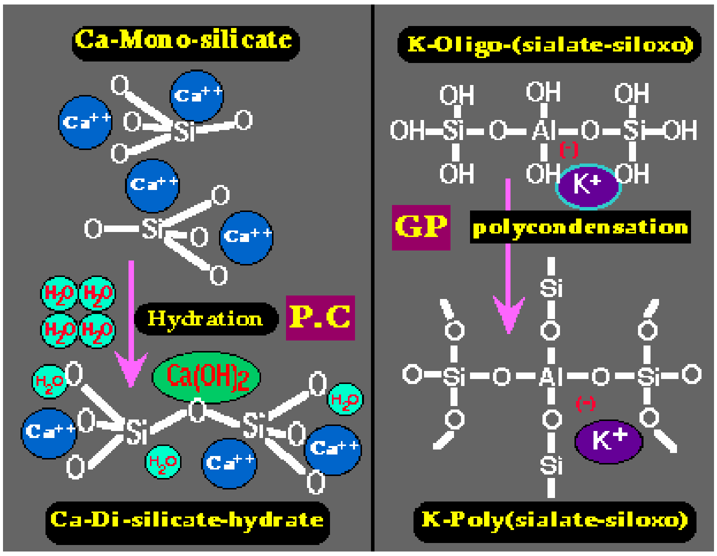

Figure 1: Comparison between Portland cement (P.C) and Geopolymer (GP)

GEOPOLYMER® is made of two components: a powder component consisting of meta-caolin and blast furnace slag, and liquid Potassium Silicate (water glass). GEOPOLYMER® sets rapidly at room temperature and provides compressive strengths in the range of 20 MPa, after only 4 hours at 20°C, when tested in accordance with the standards applied to hydraulic binder mortars. The final 28-day compressive strength is in the range of 70-100 MPa. The behaviour of geopolymeric binders are similar to that of zeolites and feldspathoids; they immobilize hazardous materials within the geopolymeric matrix, and act as a binder to convert semi-solid wastes into solids. Their unique properties which include high early strength, low shrinkage, freeze-thaw and wet-dry resistance, sulphate and corrosion resistance, make them ideal for long term containment in surface and subsurface disposal facilities. These high-alkali cements do not generate any Alkali-Aggregate reaction which adds to their favourable long-term stability.

In some aspect, solidification of hazardous residues by GEOPOLYMER® is comparable to vitrification. Like vitrification, the Geopolytec process offers:

- high strength,
- acid resistance,
- long term durability, and
- geological analogues.

But unlike vitrification, it does not require energy-consuming drying and melting. GEOPOLYTEC® requires only simple mixing equipment (such as for concrete solidification) and room temperature to harden. In fact, the close similarity of the process with common Portland cement solidification (with which it has nothing else in common, though) and its simplicity make the GEOPOLYMER® an attractive alternative to more expensive high-tech solidification methods.

Theoretically, mixing sludges directly with GEOPOLYMER® would be enough for solidification. However, in practice, a two-step technology has been elaborated having some advantages which are described in detail in (5):

1. producing a granular pre-product of the sludge and ordinary Portland cement which can be stored some days
2. binding the granular pre-product with GEOPOLYMER® and filling the resulting end-product to Big Bags or moulds.

The "inflation factor" (i.e. the volume of the solidified blocks compared to that of the sludges) is about 1.8 but can vary largely depending on the wastes to be solidified.

In order to obtain regulatory permission to employ the process in the solidification of radioactive sludges which can then be released from regulatory control, a wide range of tests was performed which aimed at demonstrating that

- GEOPOLYMER® had unambiguously formed in the process (thus precluding the formation of spurious mineral compounds)
- the solidification product shows the same properties of long-term stability and stress resistance as the pure binder matrix.

Parameters tested and standards used included

- Compressive strength according to DIN 18136
- Hydraulic conductivity according to DIN 18130
- Leaching behaviour both in a 24 hours standard test acc. to DIN 38414-S4 and a sequential leaching series according to ANS 16.1 (slightly adapted)
- structural stability and resistance of mechanical and chemical parameters under freeze-thaw and wet-dry cycles according to ASTM 4842(4843
- microbial stability according to ASTM G21(22 and DIN 53739
- surface structure (occurrence of fissures and fine-grained ablations)

A number of pilot and large scale demonstrations have been carried out in which NORM wastes from municipal waterworks and industrial sources such as demercurization residues have been solidified. As a result, the technology has gained acceptance by regulators and certification agencies that it can now be used as a means to release NORM wastes from regulatory control (8). This means for industrial users that their wastes can be stored in specifically prepared parts of municipal landfills instead of expensive landfills for hazardous waste, and that the administrative effort which is inevitable in the handling and disposing of radioactive wastes is reduced to a reasonable level.

6 DISPOSAL SITES AT WISMUT

There are several possible solutions for the disposal of higher radioactively contaminated residues at WISMUT. Disposal locations at WISMUT include:

- underground disposal in a mine adit,
- disposal in beach areas of tailings ponds,
- disposal in mine dumps,
- disposal in a contaminated mixed waste landfill.

All above ground engineered facilities were constructed following design guidelines for hazardous waste landfills. They are equipped with base sealing and drainage systems to drain off infiltration waters. An optimised system of quality assurance and monitoring serves to ensure and monitor long-term stability of residue disposal. Licensing of the WISMUT disposal sites involved comprehensive environmental impact studies, e.g. on

- contaminant release from the landfill body over time, and on
- relevant exposure pathways taking working conditions and site-specific scenarios into account.

Exposure estimates for complex situations were carried out using the recently developed software package DosModBerg®. The results of the exposure estimates confirmed that all disposal sites safely comply with the 1 mSv(a requirement according to the regulations which are to be applied at WISMUT. Hence, they also comply with the requirement of the new German Radiation Protection Ordinance for the reuse or disposal of residues.

The disposal location on waste dump 371 at the Aue site is one example that demonstrates the technical solution agreed upon by WISMUT and the permitting agencies for the disposal of water treatment residues. Figure 2 illustrates the layered structure of the facility and exemplifies measures taken to minimise contaminant release from the landfill body (base sealing, drainage system, final cover). Figure 3 is a view of the facility.

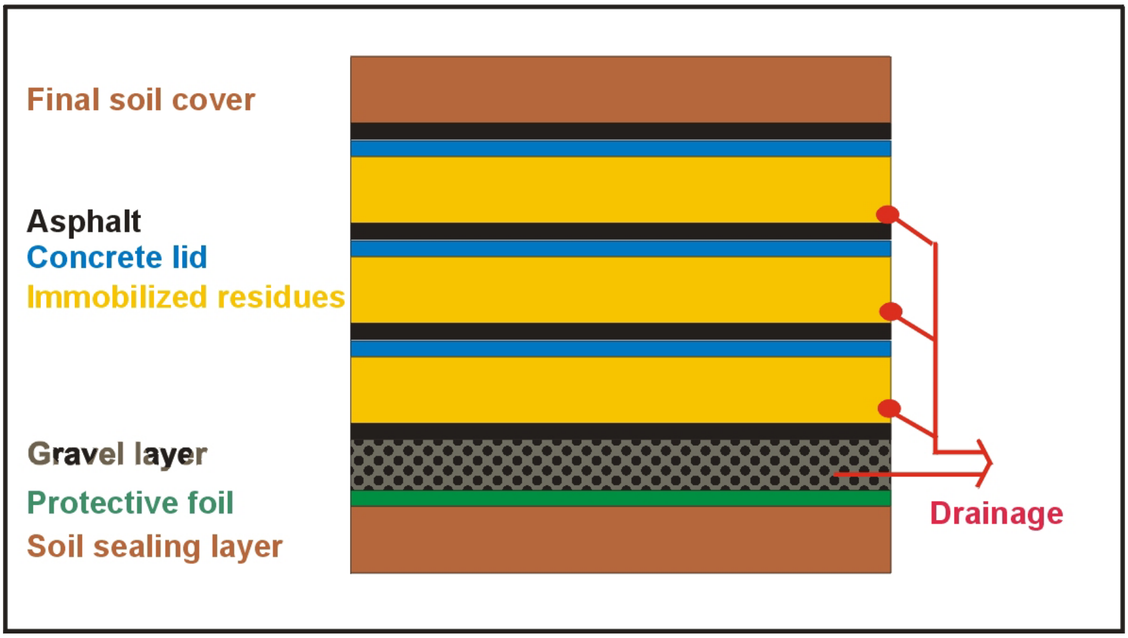

Fig. 2: Layered structure of the disposal facility for residues from the Aue WTP

Fig. 3: Disposal site of residues from the Aue WTP

7 CONCLUSIONS

Given the stringent regulatory requirements for immobilisation and disposal of water treatment residues at WISMUT, the approved technologies and licensed sites are regarded as suitable for the disposal of NORM with elevated specific activity. The GEOPOLYTEC® technology allows to achieve a degree of immobilisation by which most of the NORM immobilised by this technique will qualify for placement into special waste landfills. Large quantities of residues with elevated specific activities might be suitably disposed of at engineered disposal sites constructed for water treatment residues at WISMUT.

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