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CONCERTED ACTION TO EVALUATE THE CURRENT STATUS OF NORM - MATERIAL MANAGEMENT IN NON - NUCLEAR INDUSTRIES OF THE EUROPEAN MEMBER STATES

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INTRODUCTION

In 1996, the European Commission adopted the revised Basic Safety Standards in which radionuclide specific exemption levels are submitted (1). For radionuclides from natural origin, these exemption levels are more restrictive than the existing regulations. Industrial processes can enhance the concentration of radionuclides in waste streams which may lead to significant radiation protection problems. To identify possibly influenced industries the EC set up expert working groups and started concerted actions. One group investigated the need and the influence of the new criteria to waste management in non - nuclear industries. To gather information about treatment methods, a questionnaire was started and sent to about 190 addressees in 22 countries from which 12 are EC member states. The response and results are summarized in the paper. The work has been carried out under a concerted action contract (N° F14W-CT98-0042) with the European Commission in the framework of its 4th R & D programme ‚Nuclear Fission Safety‘ (2).

CONCERTED ACTION „IMMOBILIZATION AND SEPARATION“

The working group represents related industries and consultants dealing with NORM waste. as there are

- phosphate rock processing industry,
- natural gas and oil industry,
- steel production industry,
- zirconium sand industry,
- waste management services companies and
- consultants of industry and authorities.

The group consisted of eight representatives from the following companies / institutes :

Research Institutes
- IBR Consult B. V., Haelen, NL, coordinator
- Nuclear Research and Consultancy Group Arnhem, NL
- Brenk Systemplanung, Aachen, D

Service Companies
- Siempelkamp Nuklear- und Umwelttechnik GmbH & Co., Krefeld, D

Industries involved with NORM
- Thermphos International B.V., Vlissingen, NL
- Nederlandse Aardolie Maatschappij B. V., Assen, NL
- Hoogovens Staal B. V., Ijmuiden, NL
- Ankerpoort N.V., Geertruidenberg, NL

The work schedule, extended over 12 months, was divided into three phases :

1. Phase : Collection and evaluation of the treatment processes.

2. Phase : Inventory of the problems associated with NORM.

3. Phase : Recommendations and R & D needed for the treatment of wastes with enhanced levels of natural radionuclides.

In a workshop the results were presented and discussed with a group of invited experts from several EC member states.

GENERAL APPROACH CONCERNING IMMOBILIZATION AND SEPARATION

Management of radioactive waste from the nuclear industry is very expensive due to the high activity inventories, the long life time of the radionuclides and the required radiation protection measures. For the large amount of waste generated in non - nuclear processes with low - level radionuclide concentrations, immobilization of the radionuclides in a solid matrix is aimed at to facilitate storage in alternative waste repositories. Furthermore, the large waste volumes can be reduced by separation technology resulting in separated waste parts consisting of a higher level of radioactivity and a material free of radioactivity and therefore reusable. The availability of technologies for separation and immobilization is essential for this strategy. A questionnaire was sent to related industries to gain information about the local evaluation of this situation.

QUESTIONNAIRE RESULTS

The questionnaire was subdivided in three sections :

- general questions
  which material? ; which nuclides? ; which handling? ; which treatment?

- immobilization
  which technique? ; legislation / regulation considered? ; specification? ; economics?

- separation
  which technique? ; options for the „clean“ part and remaining part? ; legislation / regulation considered? ; economics?

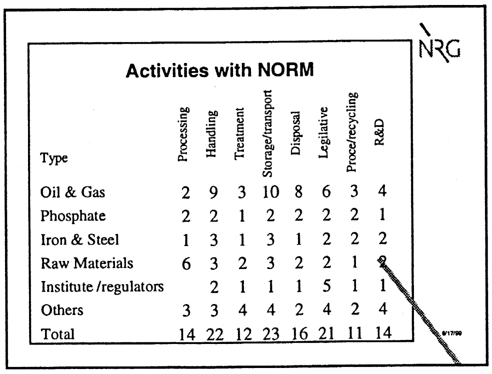
The evaluation based on the response of 64 from 192 mailings is given in the charts of table 1.

The involved amounts of NORM as well as the concentrations are in a wide range within each branch. The oil and gas industry are dealing with small amounts of sludges. The thermal phosphate industry handles phosphate ore, scales, slags and calcinate with NORM. The chemical phosphate industry handles scales and large quantities of gypsum with slightly enhanced levels of radionuclides. In the iron and steel industry waste has to be managed as sludges, residuals and blast furnace slags. Fly ashes from coal combustion have nuclide concentrations below 0.2 Bq/g. NORM wastes are often contaminated with other hazardous components like mercury, heavy metals, $BaSO_4$ or hydrocarbons which is more important for the selection of the treatment technology than the radioactivity.

With view to immobilization and / or separation techniques for their specific type of NORM waste, immobilization is not seen as a feasible option in many cases whereas different separation techniques are being used in all branches.

High pressure water jet is the favourable cleaning technique for NORM contaminated equipment in the oil and gas industry. When the efficiency is not high enough, melting of NORM contaminated scrap results in full separation of the radioactivity from the metal to slag and filter dust. Also a chemical decontamination method has been considered. Separation leads to clean material for reuse and remaining waste to be stored; repositories for this kind of waste are not available for the time being, but should be installed in the EC member states in due time. Depending on the residual waste activity and kind of waste options such as landfill, recycling or disposal as radioactive waste, may be considered.

Table 1 : Evaluation of questionnaire results



Activities with NORM

Type	Processing	Handling	Treatment	Storage/transport	Disposal	Legilative	Proce/recycling	R&D
Oil & Gas	2	9	3	10	8	6	3	4
Phosphate	2	2	1	2	2	2	2	1
Iron & Steel	1	3	1	3	1	2	2	2
Raw Materials	6	3	2	3	2	2	1	2
Institute /regulators		2	1	1	1	5	1	1
Others	3	3	4	4	2	4	2	4
Total	14	22	12	23	16	21	11	14

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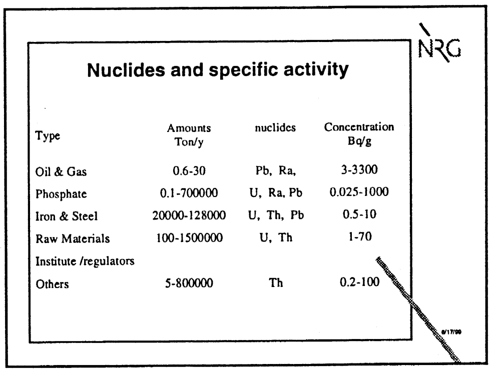

Nuclides and specific activity

Type	Amounts Ton/y	nuclides	Concentration Bq/g
Oil & Gas	0.6-30	Pb, Ra,	3-3300
Phosphate	0.1-700000	U, Ra, Pb	0.025-1000
Iron & Steel	20000-128000	U, Th, Pb	0.5-10
Raw Materials	100-1500000	U, Th	1-70
Institute /regulators			
Others	5-800000	Th	0.2-100

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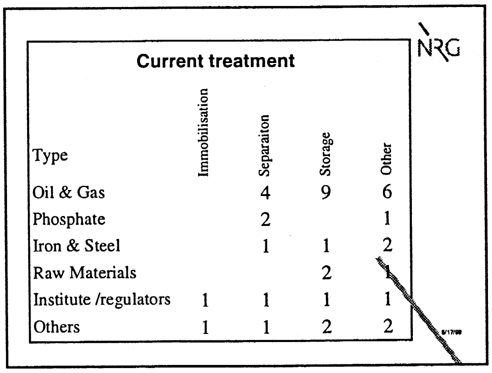

Current treatment

Type	Immobilisation	Separaiton	Storage	Other
Oil & Gas		4	9	6
Phosphate		2		1
Iron & Steel		1	1	2
Raw Materials			2	1
Institute /regulators	1	1	1	1
Others	1	1	2	2

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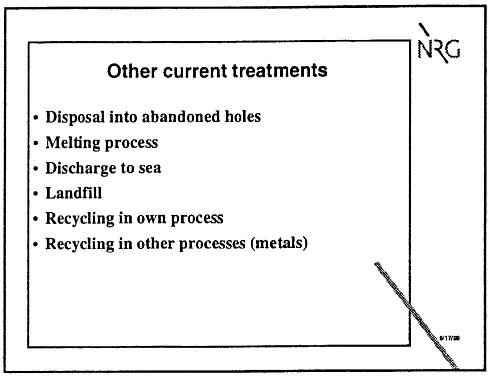

Other current treatments

- Disposal into abandoned holes
- Melting process
- Discharge to sea
- Landfill
- Recycling in own process
- Recycling in other processes (metals)

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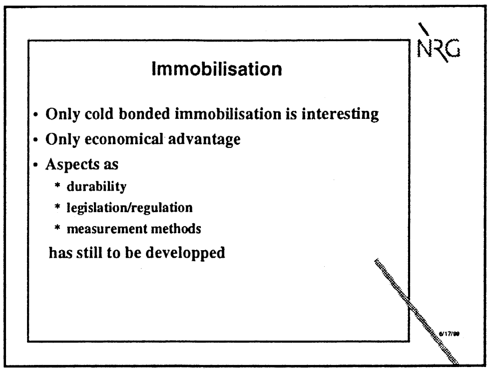

Immobilisation

- Only cold bonded immobilisation is interesting
- Only economical advantage
- Aspects as
  - * durability
  - * legislation/regulation
  - * measurement methods

  has still to be developped

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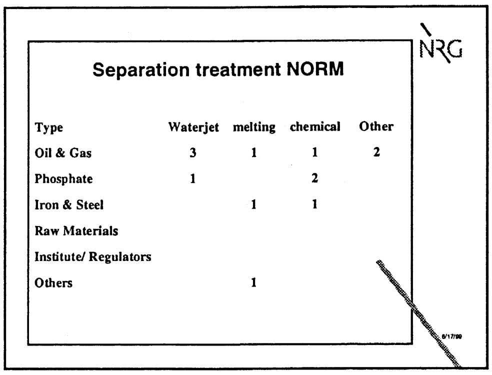

Separation treatment NORM

Type	Waterjet	melting	chemical	Other
Oil & Gas	3	1	1	2
Phosphate	1		2	
Iron & Steel		1	1	
Raw Materials				
Institute/ Regulators				
Others		1		

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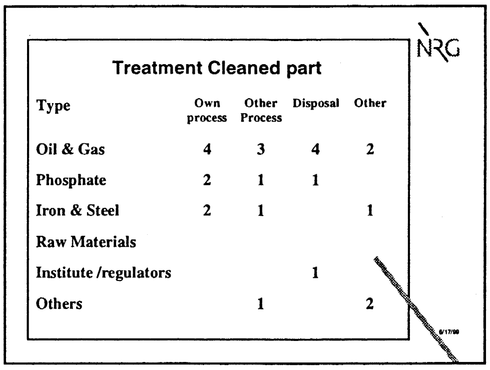

Treatment Cleaned part

Type	Own process	Other Process	Disposal	Other
Oil & Gas	4	3	4	2
Phosphate	2	1	1	
Iron & Steel	2	1		1
Raw Materials				
Institute /regulators			1	
Others		1		2

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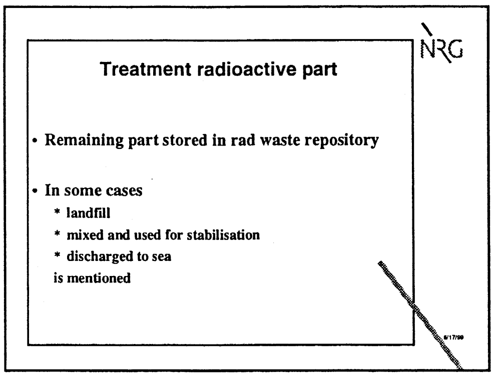

Treatment radioactive part

- Remaining part stored in rad waste repository

- In some cases
  - * landfill
  - * mixed and used for stabilisation
  - * discharged to sea

  is mentioned

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IMMOBILIZATION OF SLUDGES FROM A SINTERING PLANT

In the steel industry, investigations for immobilization of sludges contaminated by Pb 210, Bi 210 and Po 210 with an averaged activity range of 60 Bq/g have been performed.

In modern iron making, iron ore is agglomerated prior to being fed into the blast furnaces. These agglomerates are either produced in a sintering plant, or pellets produced in a pelletizing plant. The iron burden fed to the blast furnaces consists of 50 % sintered and 50 % pellets. Both agglomerates are produced on site in plants located at the integrated iron and steel - making facilities.

In the process of sintering, a mixture of iron ores and additives (like lime) is baked together at high temperatures (1.300° C) to form agglomerates. Fine coke is used as a fuel to generate the necessary heat. During the sintering process, dust is emitted from the mixture, whereas most of the sulphur ends up as SO_2 in the off - gas. The dust contains iron oxides, but also heavy metals and radionuclides. The radionuclides involved here are the volatile metals from the tail end of the decay chain of uranium - 238 : ^{210}Pb (τ = 22.3 years), ^{210}Bi (τ = 5 days) and ^{210}Po (τ = 138 days).

The sintering plant is equipped with a wet scrubber that removes SO_2 and dust from the flue gas. The scrubber water is cleaned in a water treatment plant. Here the dust, including the heavy metals and radionuclides, is remaining behind as two separate sludge fractions, named sludge # 1 and sludge # 2. Sludge # 1 (some 1500 tons / year on a dry basis) was originally taken out of the stream and stored separately. Later, it was decided that this sludge can also be recycled in the sintering plant. This is still going on today and no process problems have been encountered so far.

As alternative, two cold immobilization techniques for the sludge have been developed in a R & D stage. In process A the sludge is mixed with organic resins and cement to form a concrete - like product suitable as building material. When the immobilized sludge is applied as a covering layer for waste repositories, a first indication of the cost would be 60 - 80 Euro / t. This would be less expensive than bentonite. However, the amount of sludge available for this application is only some 700 tons per year, giving some 1500 tons per year of immobilized material. Compared to the volumes needed for this market, this amount is too small.

In process B sludge is mixed with cement or lime. This mixture is pelletized on a rotating disk. After curing the pellets over 48 hours they can substitute natural gravel in road construction. No studies have been done to determine the cost of the pellets produced from the sintering plant sludge, applied as natural gravel substitutes in asphalt. However, an estimation can be made. For a full scale plant that produces 300.000 tons of fly ash pellets per year, the costs amount to some 20 Euro / t. For the sintering plant sludge, 14 % of cement have to be added, giving an extra cost of about 8 Euro / t. Total production costs of the pellets made from the sludge will thus amount to some 30 Euro / t.

The amount of sludge available for this application is only some 700 tons per year, giving some 800 - 850 tons of pellets per year. Compared to the volumes needed for the asphalt market, this amount is too small. Therefore, there is very small economic incentive for applying the pellets in this market.

The only possibility for an economically feasible application of the immobilized sludges would be to blend the sludge with a (much) larger volume of a neutral feed stock stream, or to combine it with one or several other waste materials.

SEPARATION OF CALCINED DUST IN A PHOSPHORUS PRODUCTION PLANT

The production plant at TIBV (thermPhos International B. V. , Netherlands) consists mainly of the following stages :

- sintering plant
- phosphorus plant

In the sintering plant milled phosphate ore is pelletized with clay into granules. These granules are sintered at temperatures up to about 800° C.

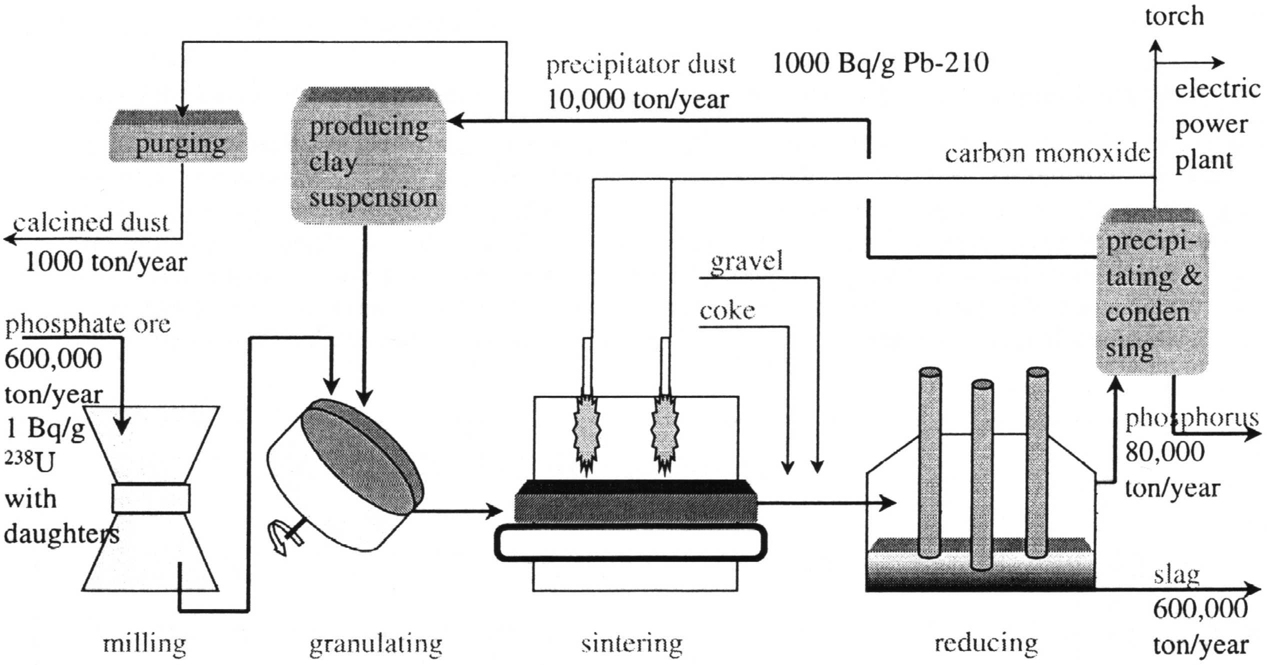
There are three electrothermal phosphorus furnaces in the phosphorus plant. The pellets are fed into the furnaces together with gravel and coke. At temperatures exceeding 1500° C a reaction takes place, in which phosphate ore is reduced to elemental phosphorus (P_4)

A calcium silicate slag is formed which flows continuously from the furnace. The elemental phosphorus leaves the furnace as a gas, together with the carbon monoxide formed during the reaction. Entrained dust is separated from the gases in an electrostatic precipitator. This dust is collected in the slurry tanks, where it is mixed with water (Figure 1).

Specific radioactivity content of the NORM material

The phosphate ore contains approximately 1 Bq/g of ^{238}U. All daughters of the ^{238}U decay chain are in a state of (approximate) equilibrium with the parent nuclide. Due to enrichment during the process the precipitator dust is enriched up to 800 Bq/g for ^{210}Pb.

Figure 1 : Process scheme of TIBV production plant.



$$2Ca_3(PO_4)_2 + 6SiO_2 + 10C \Rightarrow 6CaSiO_3 + 10CO + P_4$$

phosphate ore and gravel and coke => slag and carbon monoxide and phosphorus

Precipitator dust processing plant

In order to ensure the stable operation of the furnace process, some of the precipitator dust must be removed from the precipitator dust cycle. This precipitator dust is processed into calcined dust and is stored on site as radioactive waste. At the present time approximately 750 tons of precipitator dust are taken out of the process each year. Because storage of calcined dust is very expensive, only the absolutely necessary amount of precipitator dust is removed.

In the period 1990 - 1994 a pilot plant trial was carried out to determine whether the stored calcined dust and the precipitator dust could be chemically processed. On the basis of this pilot plant, TIBV worked out plans for an industrial - scale precipitator dust and calcined dust processing plant (referred to below as precipitator dust processing plant). In this plant the heavy metals, the radioactive fraction and the other substances are chemically separated from each other. A highly active lead sulphate fraction is formed which has to be transported to COVRA (Centrale Organisatie voor Radioactief Afval : The Dutch central organisation storing radioactive waste).

Advantages of the plant

The volume of radioactive waste would be reduced considerably (see under „effectiveness“). Due to the above reduction, it would be possible to remove more precipitator dust from the process. The emission of radionuclides is reduced by a factor of about 2 if, instead of 1,000 tons per year, 3,000 tons of dust per year were removed and processed in the precipitator dust processing plant. The dose level in the surrounding area as a consequence of the emissions would also be decreased by a factor of 2.

Effectiveness

It cannot be excluded that the activity of the separated zinc fraction (150 tons per year with an expected activity of 30 Bq/g of ^{210}Pb) will be above the future limiting value of 10 Bq / g (1), and therefore be regarded as a radioactive waste. The most important objective of the project - concentration of activity - would then be only partly realized.

Economical aspects chemical separation

Formally, no criteria for ALARA analyses exist to weigh the effects of measures on the reduction of the dose in the surrounding area (in this case by roughly half) such as implementation of a precipitator dust processing plant against the investment costs of 14 million Euro for a full - scale plant and the operating costs of 3 million Euro / year over 25 years. The Dutch policy statements give no clear indication of what costs are adequate in relation to the reduction of individual or collective dose. Due to the lack of nationally or internationally accepted values, TIBV referred to a CBA (Cost Benefit Analysis) value for the purpose of weighing up the reasonableness of an investment regarding the reduction of the collective dose. The figures published by the National Radiation Protection Board in 1993 specify 35,000 Euro per man Sv for the general public (Doc. NRPB, 4 No. 275-80 1993). TIBV therefore applied the NRPB Cost Benefit Analysis value (CBA) of 35,000 Euro per saved man - sievert in order to weigh up the reasonableness of an investment.

Precipitator dust processing plant

The collective dose caused by TIBV is 2 man Sv per year. Based on an investment for 25 years, the collective dose reduction can be calculated to :
2 [man Sv per year] * 25 [years] / 2 [reduction] = 25 [man Sv].
Applying the abovementioned CBA values, an investment of 25 [man Sv] * 35,000 [Euro per man Sv] = 0.9 million Euro, can be regarded as defensible.

The investment required to build the precipitator dust processing plant is 14 million Euro. The operating costs would be 25 years * 3 million Euro per year = 75 million Euro, so that the total costs over 25 years would amount to 98 million Euro. This is much higher than the sum NRPB is regarding as reasonable for the reduction of the collective dose. The necessity for building a precipitator dust processing plant in order to reduce the dose in the surrounding area is therefore not given. Consequently, in early 1993, after consultation with VROM (Dutch environmental Ministry), it was decided not to build up the plant.

SEPARATION OF NORM MATERIAL BY MELTING

Melting of slightly radioactively contaminated metals for recycling to products used in the nuclear cycle is an approved waste management path in Germany. About 14,000 t of scrap have been recycled in the last two decades. This waste management has to be done under The German Radiation Protection Ordinance. Lessons learnt in the past could widely be used for the melting of NORM - contaminated metals from the non - nuclear industry.

In January 1998 a second melting plant specialized to melt NORM- and / or chemically contaminated scrap started operation [3]. Technical data of the melting plants are compared in table 2. Both plants are under operation of Siempelkamp Nuklear- und Umwelttechnik GmbH & Co. at Krefeld site.

Table 2 : Technical data of the melting plants

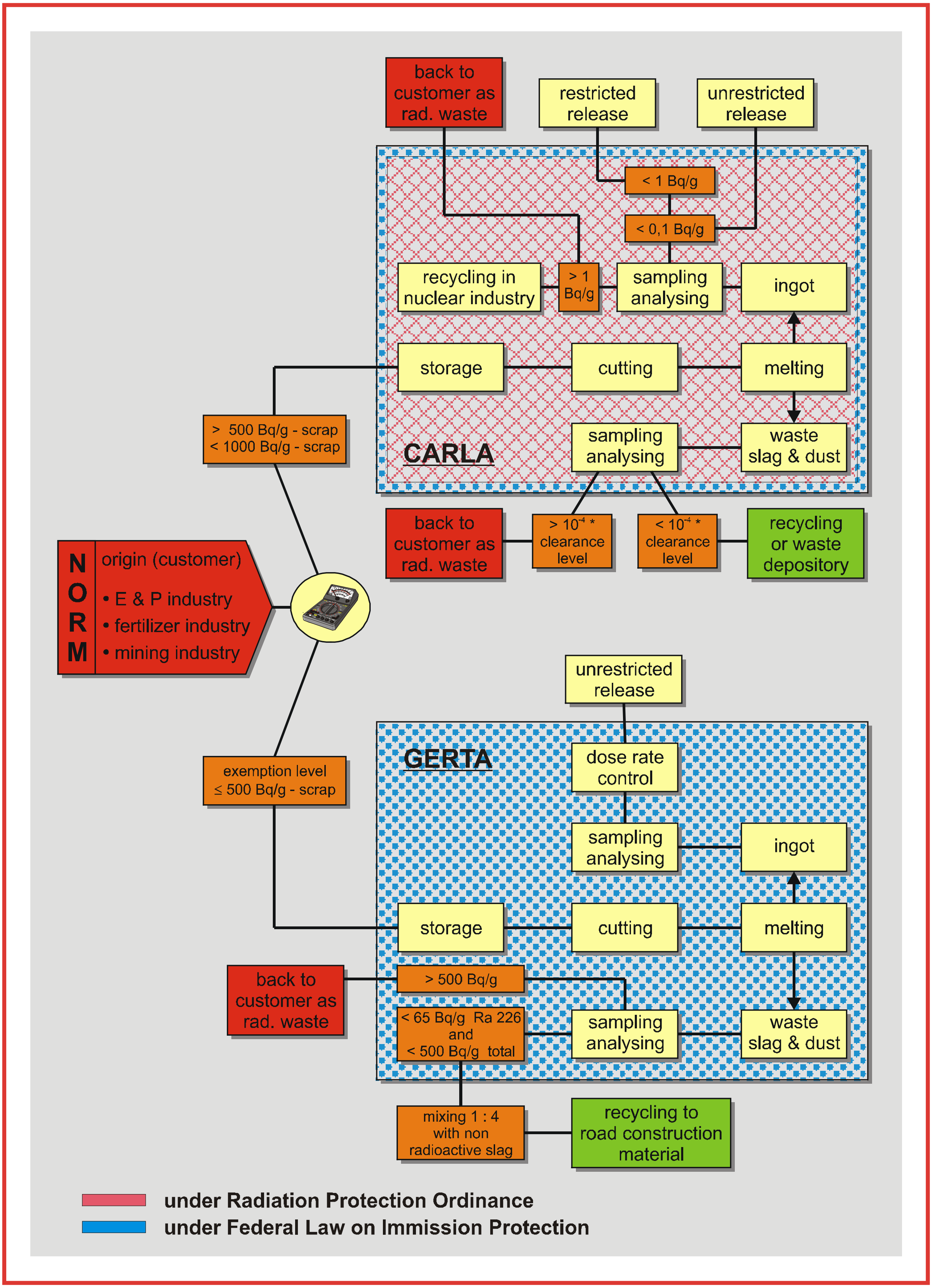
Plant		**CARLA**	**GERTA**
Starting date of operation	m / y	10 / 89	01 / 98
Annual capacity	t / y	4,000 (2 shifts)	2,000 (1 shift)
Licensing		German Radiation Protection Ordinance and BImSchG	Federal law on emission protection (BImSchG)
Limits for NORM scrap	Bq/g	1,000	500
Melting unit		3.2 t medium frequency induction furnace	8 t line frequency induction furnace
Materials		Ferro- and non - ferro metals	Ferro – metals

A flow chart showing the management and treatment steps related to NORM to be molten in the Siempelkamp melting plants is presented in Figure 2.

By melting NORM - scrap, the radionuclides like Ra 226, Ra 228, Po 210 and Pb 210 show different behaviours. Radium is concentrated in the slag, while polonium and lead go into the gas phase to be filtered with the dust. The molten metal is free of radioactivity usable as secondary raw material. The waste arising from the melting process as slag can be recycled to road construction material, if the specific activity is below 65 Bq / g Ra 226 and below 500 Bq / g in total. Filter dust is often co - contaminated by mercury or other heavy metals. In this case the filter dust is going to an underground repository. When the limits are exceeded, the waste streams have to be managed as radioactive waste. When the Basic Safety Standard is transferred into national regulations exemption and clearance levels will change. An application to NORM may increase the volume of waste produced by the non - nuclear industry.

From the economical point the important advantage of melting is the reduction of the radioactive waste volume and recovering of usable material. The costs of melting are strongly depending of the necessity of radiation protection controlling. Therefore, the specific prices for melting under full radiation protection control is in the range of 2,500 EURO / t up to 5,000 EURO / t depending on the kind of material, the kind of contamination and the total delivered mass. Treatment in an uncontrolled melting plant is cost saving and in the range of 600 - 900 EURO / t.

Figure 2 :


back to customer as rad. waste
restricted release
unrestricted release
< 1 Bq/g
< 0,1 Bq/g
recycling in nuclear industry
> 1 Bq/g
sampling analysing
ingot
storage
cutting
melting
> 500 Bq/g - scrap
< 1000 Bq/g - scrap
CARLA
sampling analysing
waste slag & dust
back to customer as rad. waste
> 10^{-4} * clearance level
< 10^{-4} * clearance level
recycling or waste depository
NORM
origin (customer)
• E & P industry
• fertilizer industry
• mining industry
unrestricted release
GERTA
dose rate control
exemption level
≤ 500 Bq/g - scrap
sampling analysing
ingot
storage
cutting
melting
back to customer as rad. waste
> 500 Bq/g
< 65 Bq/g Ra 226 and < 500 Bq/g total
sampling analysing
waste slag & dust
mixing 1 : 4 with non radioactive slag
recycling to road construction material
under Radiation Protection Ordinance
under Federal Law on Immission Protection

CONCLUSION

Results from the questionnaire show that involved non - nuclear industries have a wide range of different NORM wastes. Waste streams and nuclide inventory show a large variation. Waste treatment technologies are available. Immobilization technique is not widely used until now, because no attractive recycling market could be found for the immobilized waste. However, it was pointed out that immobilization could be an interesting option as a treatment of the radioactive remainder from separation processes. Separation is being applied in a wide range, different technologies are available. From an economical point of view the investment in new technologies is sometimes assessed as not sufficiently attractive.

Presently, official repositories and landfills are not available. Their installment should be one of the most important goals for the near future.

To exchange experiences with the application of the Basic Safety Standard (1) in the non - nuclear industry a Thematic Network will be organized in the frame of the EC Fifth Frame Programme.

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(1) Euratom (1996). Council Directive 96 / 29 Euratom of 13th May 1996 laying down the basic standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation. Off. J. of the EC L159 / 1.

(2) A feasibility study on the technical and economical aspects of treating natural radioactive waste materials (NORM)
Wiegers, IBR; Roelofs, NRG; Kugler, Brenk - Systemplanung EUR EN 1999

(3) A new recycling plant for metals, contaminated by naturally occurring radionuclides
Markus Hamm, Siempelkamp Nuklear- und Umwelttechnik, Proceedings of the NORM II - Symposium, Nov. 10 - 13th, 1998, Krefeld, Germany.