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Experience of applying regulations on industrial uses of natural radioactive materials

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EXPERIENCE OF APPLYING REGULATIONS ON INDUSTRIAL USES OF NATURAL RADIOACTIVE MATERIALS

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ABSTRACT

Natural radioactive material in Bavarian non-nuclear industry subject to regulation, is used mainly in metallurgical processing of thorium in the manufacture of tungsten electrodes for discharge lamps and welding. Additional minor applications are uranium paints in the glass-industry and radium self-luminous light-sources. Regulatory measures such as license conditions and inspections concentrate on safe handling of the material on one side and appropriate dosimetry on the other. Workplace monitoring is performed by neutron activation analysis of the airborne dust. Individual personal exposure of the workers, however, can only be assessed with sufficient accuracy by alpha spectroscopy of urine and faeces. So far doses determined were within the allowable limits. On the other hand applications at present exempted from being licensed and inspected were found to produce interior doses close to the limit values. Typical problems in the waste management are high disposal prices for alpha emitting material and a lack of recycling facilities for radioactive scrap metal.

INDUSTRIAL USES OF NATURAL RADIOACTIVE MATERIAL

Thorium

Due to its low photo electron ionisation energy and metallurgical and thermal properties thorium is the most effective additive to tungsten electrodes for inert gas welding as well as for high performance discharge lamps. It helps stabilising the electric arc and minimising electrode wear by sputtering.

In the last years thorium was successfully replaced by lanthanum in welding rods. For various reasons, however, establishing the new rods full scale in the welding industry is far from accomplished. Thus the annual turnover of thorium in the production of welding rods in Bavaria has risen from 315 kg to 1900 kg thorium oxide or 15 GBq Th 232/Th 228 resp. in the last 20 years. At present the increase in thorium consumption has apparently stopped and the relative share of thoriated electrodes of the total of electrodes produced is decreasing.

Despite many research and development efforts so far no equivalent for thorium has been found in lamp technology, since ThO2 containing or coated electrodes in xenon arc or metal vapour lamps are unique as far as discharge ignition, arc stabilising, lower tungsten sputtering causing less staining of the glass housing and thus longer lifetime is concerned. Approximately 1500 MBq (180 kg) of Th/W powder and Th(NO3)4 are being processed per annum in the Bavarian lamp industry.

Uranium and Radium

The addition of uranium compounds to glassware for optical or decorative reasons has decreased to a few kilograms per year, while handling of radium now concentrates on the removal and replacement by non-radioactive phosphors particularly in self-luminous aircraft instruments.

Uranium, Thorium and Progeny not controlled by Regulation

Besides these well-known and regulated uses certain industrial activities involve natural radioactive material not regulated by radiation protection legislation because the activity concentration is below the exemption level, e.g. the use of the abovementioned welding rods in TIG-welding, or the radioactive substances are an (unwanted) natural by-product and not the objective of the actual process, e.g. the manufacture of zirconium ceramics or the extraction of niobium, which exist exclusively in ores

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containing more or less uranium and thorium. Nevertheless some of these activities cause considerably high inhalation rates as well as disposal problems. Since ores or minerals are in radioactive equilibrium, the total specific activities of these substances are usually higher than in the separated species and, depending on the intensity of the contact, remarkable dose rates of workers may occur.

LEGAL ASPECTS

Exemption Levels

In Germany the basic elements such as regulations, licensing, notification/registration, inspection, and enforcement are established in the *Radiation Protection Act.* Besides general rules for the safe use of radioisotopes, a number of limits are established in order to restrict the radiation exposure to workers and the public to an acceptable minimum.

Based on their radiotoxicity all radioisotopes are classified in activity levels below which the professional use of the nuclides requires neither licensing nor registration.

For the use of chemically separated Thorium the limit is 50 kBq, i.e. 25 kBq Th 232 and 25 kBq Th 228 and in regulations usually referred to as Th nat. For Th 230, depending on the provenance normally contributing from below 10 % to 40 % to the total activity, the exemption level is 5 kBq. For uranium used in its chemical form, defined as U nat, the activity distribution is 49 % U 238 and U 234 each and 2 % U 235. The exemption level is 5 MBq.

The relevant radium isotopes like most alpha emitting nuclides are exempted below 5 kBq.

Solid state matter containing less than 500 Bq/g natural radioisotopes may be used without license or registration regardless of the total amount involved. Typical applications are the electrodes mentioned. For Th nat 500 Bq/g correspond to 6.2 % by mass whereas usual concentrations are appr. 2 - 4 %.

Dose, Emission, Disposal Limits

Under the regulations of the radiation protection act licensees have to comply with the following limits stated in the act or derived thereof in connection with ICRP recommendations.

Effective Dose (whole body, gonads, bone marrow, uterus) for occupationally exposed persons category A (workers medically and radiologically monitored)	50 mSv/a
occupationally exposed persons category B	15 mSv/a

	Th nat	U nat
Maximum annual intake by inhalation (ALI) (Bq)		
Oxides and hydroxides (Th)	100	500 -
Others (Th)	50	30000
Airborne activity concentration (DAC) (mBq/m3) in controlled/non-restricted areas for		
Oxides and Hydroxides (Th)	42/0.06	210 - / 0.3 -
Others (Th)	21/0.03	12600 / 18
Surface contamination in controlled workplaces in Bq/cm2	50	50
non-restricted areas	0.5	0.5
Activity concentration in sewage water (Bq/m3)	300	3000 - 30000
Activity concentration in effluent air (Bq/m3)	0.0001	0.05 - 1

Clearance level for disposal (Bq/g)		
Uses exempted from regulation	5	500
Draft of disposal act for licensed uses	Th 232 0.008/0.3	U 238 0.1/4
Clearance/incineration (Bq/g)	Th 228 0.05/1	U 235 0.1/3
	Th 230 0.05/1	U 234 0.1/3

RESULTS AND EXPERIENCES OF REGULATORY PROGRAMME

Authorisation and Inspection

Applicants for authorisation of radioactive practises have to provide evidence of reliability, training, technical skill and facilities available for the handling of radioactive matter at a minimum of personnel exposure as well as for monitoring purposes. Additionally conditions are formulated in the license for inspections and measurements to be carried out by external consultants. External and internal dosimetry are performed by authorised institutions only.

Apart from these regular measures the authority inspects and reviews the activities and compliance with license conditions at unannounced visits on-site. Inspection check lists comprise the control of records of medical checks, dosimetry results, contamination surveys, consumption and waste management, training. One of the objectives of on-site inspections, of course, is the review of all operations in order to discuss possibilities of improvements.

In most cases these discussions resulted in the installation of new or improved housing and ventilation of individual workplaces like presses or grinding machines, where the radioactivity concentrations exceeded the allowable values, particularly since the alternative being forced to wear dustmasks is difficult to control and not favoured by workers.

Dosimetry

Standard film badge dosimeters are used generally in controlled areas. The external doses detected monthly by this method have hardly ever been found to exceed the detection limit of 0.2 mSv. Since the critical path for alpha particles is inhalation, film badges may yield a representative personal dose only in certain cases, where dustfree handling of old material with high gamma-doserates of the radioactive Th daughter nuclides is concerned. Hence according to the *German Guideline for Physical Radiation Protection Control* methods have to be applied which allow an accurate assessment of incorporated radioactivity. Up to a few years ago whole-body counting was used, but had to be discarded, for the detection limit is more or less close to the dose limit of 50 mSv. So in the same time two other means of internal dosimetry have been developed to satisfactory performance:

Workplace Thorium Air Concentrations

The well-proven techniques of sampling airborne particulate matter on various filter types for the analysis of heavy metals and other solid dust components are equally suited for the analysis of radionuclides. For thorium isotopes the analysis may be carried out by neutron activation followed by gamma-spectroscopy of Pa 233 (NAA) or by direct alpha spectrometry (DAS) in Frisch grid ionisation chambers (Hötzl et al. 1996). While NAA is less affected by filter depths and variations in particle size, DAS is less time consuming, does not require a neutron irradiation facility and yields the additional information of all Th-isotopes. The minimum detectable activity is 2 mBq for DAS and 0.4 mBq for NAA which - for air samples of 100 m3 - is sufficient to monitor the concentration limits stated in 2.2. At present an electrostatic precipitation sampler is tested in our laboratory producing samples completely free of filter absorption interference for DAS.

Excretion Measurements

In order to investigate the long term accumulation of alpha nuclides or the short term incorporation e.g. by accidents or incidents the determination of the isotopes in urine or faeces resp. is the only method suf-ficiently sensitive and specific. The *Laboratory for Radiotoxicology* of the LfU as the official institution in Bavaria for excretion measurements has established the facilities and elaborated the standard method of sample preparation by ashing, microwave digestion, and electrodeposition prior to direct alpha spectroscopy. Detection limits for Thorium are 1 mBq/sample. The assessment of the effective doses is based on the biokinetic and dosimetric models of ICRP 54, 30, 23 and dose coefficients as published by the German Government 1989. Minimum detectable effective doses as calculated from urine analysis are < 5 mSv/a.

Results

As the excretion measurements have only been running for a relatively short period, there is no data basis yet to correlate workplace concentrations with urine or faeces excretions determined. Besides sampling for the excretion analysis on-site is still met by various practical problems, for instance overall contamination of sample containers resulting in analytical artefacts. The results obtained so far, however, indicate that in most cases workplace measurements are not representative for the effective time and local concentration of the worker's exposure. One way out of this problem would be the use of personal samplers, which on the other hand are regarded as hindrance by many workers.
Nevertheless, from the results obtained so far by all methods applied, it can be stated that doses of workers in Bavarian plants handling unsealed natural radioactive material are of the order of or below 5 -10 mSv/a. In certain operations, however, 50 mSv/a may be exceeded when protection masks are not reliably used.
A particular research project is under way by the *Laboratory of Radiotoxicology* to investigate the in-halation of thorium with the welding fumes of TIG welders. Since welders are exempted from regulatory control, the results of these urine screening project may help to either put this radioactive practice under control or to accelerate the replacement of thorium by lanthanum. So far the results indicate that doses above 5 mSv/a are likely to occur thus classifying the users as occupationally exposed persons.

CONCLUSION

The regulatory programme implemented on practices of industrial processing of natural radioactive matter like thorium and uranium has been shown to be both justified by the possibilities of marked exposure par-ticularly through thorium inhalation and successful as far as the minimisation of risks by improvements of day-to-day safety measures could be enforced. For the internal dosimetry as essential part of radiation monitoring the activity air concentrations on-site were found to give sufficient information of critical work-places, whereas effective doses related to individual workers are more accurately assessed by excretion analysis.
Apart from regulated applications, practices involving natural radioactivity under present legislation exemp-ted from control were investigated. Incorporation hazards for workers as well as uncontrolled dumping of radioactive waste implied with these occupational operations suggest that regulations made feasible by Title VII of the EU Basic Safety Standards could improve radiation safety and at the same time increase pressure to discard unnecessary uses of radioactive material.[2]

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[2]Hötzl, H.; Riedmann, W.; Weinmüller, K.; Winkler, R. Comparison of Direct Alpha Spectrometry and Neutron Activation Analysis of Aerosol Filters for Determination of Workplace Thorium Air Concentrations. Health Phys. 70:651-654; 1996