Paper 18

Incorporation of radionuclides by workers in thorium processing industries - monitoring and minimising internal doses

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Abstract

Due to the conditions prevailing in industrial workplaces and the amounts of pure thorium used in relation to the extremely low limits of intake by inhalation the thorium processing industry is among the non-nuclear users of radioisotopes with the highest radiation hazards. This report presents the regulatory and technical measures to control the incorporation of thorium in order to minimise internal doses. Various dose assessment techniques are described as well as workplace protection measures. In the practical part the difficulties of the enforcement of protection measures on site and the shortcomings and uncertainties of the dosimetry methods applied are discussed.

Introduction

In spite of intensive research and development efforts to replace thorium by other elements of similar photo electron, metallurgical and thermal properties the producers of tungsten electrodes for inert gas welding as well as for high performance discharge lamps rely largely on thorium as the most effective additive. Thus the annual turnover in Bavaria has stabilised at 1900 kg thorium oxide (15 GBq 232Th/228Th) for welding rods and 180 kg (1.5 GBq) thorium oxide or nitrate in the lamp industry. While the external radiation is easily monitored, the main radiation hazard arises from inhalation of thorium or thorium contaminated dust during the various production operations such as the large scale processes of sintering or pressing in the welding rod industry, the equivalent but smaller scales deployed in lamp technology or the use of welding rods in many industries.

Legislation and technical regulations

The use of thorium according to German legislation is subject to notification for activities exceeding 50 kBq of “Th-nat“, i.e. chemically separated thorium equivalent to 6.2 g, and subject to licensing for activities more than 500 kBq. Depending on the time elapsed since separation disequilibrium between 232Th/228Th of up to _ has to be allowed for. The general exemption limit for naturally occurring solid radioactive material is an activity concentration of 500 Bq/g irrespective of the absolute amount. When subject to licensing, conditions are to be established to set up safe operational practices. Legislation is supplemented by various prescriptive regulations such as guidelines and standards concerning training, dosimetry, shielding etc., the most important ones for users of unsealed sources being: Guideline for the physical radiation protection control - committed dose assessment. Guideline for the assessment of internal doses - basis for calculation. These guidelines establish threshold limits and criteria of practice which require incorporation monitoring and describe the monitoring techniques applicable and list the biokinetic data required for dose calculations such as excretion and retention coefficients. In accordance with these guidelines regular monitoring of incorporation is required, when the maximum incorporable activity exceeds one tenth of the respective ALL. For thorium the critical path is inhalation and the annual limit of intake is 100 Bq Th-nat for oxides / hydroxides and 50 Bq for other chemical species. To determine the inhalable activity the days of exposure and the relative portion of the workplace activity that can be inhaled unnoticedly has to be taken into account. In addition further quantitative regulations are established, e.g. detection limits for the methods to be applied of 3 % of the ALL and the requirement of further investigations by an additional method, when first results show exposures exceeding 10 % of the ALI. Thus for monitoring of inhalations of any radioactive material the determination of one or a combination of the following parameters is listed: workplace airborne thorium activity concentration thorium activity of urine or faeces exhalation of thoron body activity. While the body activity counters do not achieve the detection limits required for thorium monitoring, all other methods are being used or under investigation. Their individual merits will be discussed later in the paper.

Internal dose assessment techniques

Workplace thorium air concentration. A variety of samplers of airborne particulate matter for the analysis of dust components has been well-proven in outside atmospheres as well as in workplaces. Of particular interest in our case are high-volume samplers to yield sufficient material during short campaigns of handling and personal samplers to simulate the breathing conditions of workers in a more representative way. The analysis of thorium isotopes may be carried out by neutron activation followed by gamma spectrometry of 233Pa (NAA) or by direct alpha spectrometry (DAS) in Frisch ionisation chambers. While NAA is less affected by filter properties and variations in particle size, DAS is less time consuming, does not require a neutron irradiation facility and provides the additional information of all alpha-emitting progeny, which, as will be shown later on, is important for the interpretation of the incorporation data. The minimum detectable activities of 2 mBq (DAS) and 0.4 mBq (NAA) are sufficient to monitor even concentration limits in non-restricted areas of 0.03 mBq/m3 at least for hi-vol samples. Other analytical means are in use such as liquid scintillation after chemical preparation or - particularly fast, but without nuclide specific information - conventional x-ray fluorescence spectrometry. Excretion analysis since measurements of air activity concentrations produce merely the information of the amount of inhalable thorium with the amount of actually inhaled matter being subject to further assumptions on the worker's behaviour, the excretion of thorium represents the actual body burden of radioactivity. At least in theory: the concentrations determined in urine or faeces have to be converted to committed dose equivalents or inhaled activities in order to check compliance with dose limits or ALIs. These calculations again are based on the assumptions of dosimetric and biokinetic models describing the metabolism of radioactive elements and their retention in specific organs and tissues as well as their excretion over time. The data used in the German guidelines are ICRP publications 23, 30, 54, but more sophisticated 14-compartment models and adapted incorporation and inhalation dose coefficients have been developed in ICRP 66, 67, 69, 71 and reevaluations of the committed dose equivalent estimations have been suggested (Lee et al. 1997), (Noßke 1998). It is generally believed that more recent models and dose coefficients lead to lower and more realistic estimates of doses. According to the models used at present daily excretion for chronic inhalation of 1 Bq/d of 1-micron-particles 232Th is 0.4 mBq/d in urine for the first days increasing to 5 mBq/d after 4000 days, and 54 to 600 mBq/d resp. in faeces. After an acute intake the urinary excretion will be 0.3 mBq/d for 1 Bq inhaled activity, but decreases rapidly by a daily factor of 4 to 2 during the first days and stabilises after 10 days at appr. 0,003 mBq/d. Faecal excretion under these conditions is from 54 to 160 mBq/d the first 3 days and then drops sharply to less than 1 mBq/d after 10 days. The standard analysis method of sample preparation by ashing, microwave digestion and electrode position prior to direct (-spectrometry elaborated in the Laboratory for Radiotoxicology of the Bavarian State Office for Environmental Protection achieves detection limits of 1 mBq per sample for thorium isotopes. As this method provides all isotopic information of the decay series, allowance for the background incorporation of 228Th via 228Ra ingestion can be made judged on the excess activity of 232Th expected from occupational exposure by means of the 232Th/228Th ratio of the material determined in vitro. So, based on these facts and assumptions, industrial operations and workplace conditions are inspected and evaluated and – considering the dominant way of intake being chronic or rather in campaign intervals - a monitoring programme is set up with the company's management and RSO. Thoron (220Rn) Exhalation Measurement Thoron in breath measurements have been in use for years particularly in thorotrast therapy. Research carried out at the Federal Office for Radiation Control, Berlin, (Eisenmenger 1998), with the objective of developing a routine technique with detection limits required for workplace monitoring has shown that electrostatic precipitation of the thoron daughters prior to the (-spectrometric determination of particularly 212Po is capable to detect 228Th in the lungs at a level of 3 % of the ALI corresponding to 6 Bq of inhaled 228Th . So far, however, no interpretation model has been agreed on as to the exhalation rates in relation to the 232Th body content. This uncertainty as well as lack of information on the 232Th/228Th ratio renders dose estimations and the discrimination between naturally (background) and occupationally incorporated Thorium inaccurate. Nevertheless with regard to the tedious efforts of excretion analysis and practical problems affecting the accuracy of this technique, as reported below, the relatively rapid and simple exhalation technique offers the advantage of more frequent measurement series, thus improving representativity and statistics under fluctuating conditions.

Experience and results of control programme

Control of workplace atmosphere intercomparison sampling and activity concentration measurements by means of different samplers and spectrometric methods resulted in good agreement within about 20 % rel. only when samples were taken not to close to the operation concerned. For workers directly linked with and therefore adjacent to the dust emission, airborne thorium concentrations determined on filters taken with personal samplers showed differences of more than a factor of 2 within samplers carried by the same person because of the dust inhomogenity. When using (-spectrometry, the age of the material has to be considered diligently with respect to the nuclides analysed: especially 228Ac reaches only 10 % of the 228Th activity 1 year and 20 % 2 years after separation, which is not an unusual situation for users regularly supplied. Generally 224Ra or 212Bi/Pb are less prone to underestimations, since they are in equilibrium with 228Th rather fast and losses by way of thoron emanation are relatively acceptable in that kind of material. Depending on the type of industrial production and specific operational process such as sintering, pressing, grinding of wires or rods or mixing and filling powders, concentrations of Th-nat obtained were in the range of less than 1 mBq/m3 - 2500 mBq/m3, the latter ones arising from filling thorium/tungsten powder in the tubes used for pressing welding rods. The concentration limit for occupationally exposed persons with 50 mSv annual dose limit is 42 mBq/m3 for a 2000 working hours year. Excretion m onitoring from the relatively small amounts of thorium used in lamp technology up to the over 100 kg processed monthly for welding electrodes there is no continuous handling all the year round for production purposes. Usual practice is more or less regular intervals of concentrated processing of thorium followed by intervals of using other additives or producing pure tungsten components. On the other hand welders used to thoriated electrodes would be exposed to thorium dust and fumes more or less continuously. Thus it was decided to sample and analyse faeces for production workers immediately after periods of processing thorium because of the higher concentrations in the samples, and to take advantage of the much easier handling of the urinary excretion method for welders. Due to the rapid decrease of excretion after acute intake the accurate accountance of time and period of use by the workers is of decisive significance. Results obtained so far - after 3 series in the welding rods and 1 in the discharge lamps factory - ranged from faecal excretions of 10 mBq/day 228Th and 232Th - 1600/2500 mBq/day 228Th/232Th resp. A conservative estimate of the annual dose for the highest value would result in an annual dose of appr. 500 mSv. A critical review of all the results, however, reveals more or less all problems inherent to the system: As the sample receptacles are run through the complete process of analytical preparation together with the samples outside contamination of the vessels has a strong likelihood for artefacts as proven for the 500 mSv. Ingested thorium via resuspension in the saliva of exhaled thorium and through careless handling may contribute significantly to the thorium determined, but should undergo a different dose calculation; in particular the ALI of ingested thorium is 300 times higher than the corresponding value for inhalation. Although the background values exhibit always a 232Th/228Th relation well below 1 compared to industrially used thorium with ratios markedly exceeding 1 for more than 20 years after separation, it is difficult to discern occupational enrichment of 232Th from strongly varying backgrounds, particularly when dealing with workers with years of thorium history and their body depots. In order to correlate inhalation doses of a defined period of exposure to working conditions, it is therefore necessary to analyse samples immediately before and after such an operation, which certainly increases cost and workers` unwillingness to cooperate. On top of the practical problems there remain interpretation and estimation uncertainties due to regulatory differences, e.g. the presently established ALI of 100 Bq thorium oxides and annual dose limit of 50 mSv whereas by assessment by means of official guidelines or ICRP models 100 Bq would correspond to 20 mSv.

Comparably consistent dose conditions, however, were found for TIG welders in a research project carried out at the Bavarian Radiotoxicology Laboratory mentioned above (Sternad 1998). According to the urinary excretion all 30 welders under investigation were found having accumulated thorium corresponding to committed annual doses of 0.7 - 7,6 mSv with a background level of 0.5 mSv.

Measures taken on-site to minimise doses

Working conditions in thorium processing plants have long been known to give rise to remarkable external and internal doses, for airborne activities have been measured long before other internal dose monitoring techniques were developed. Thus over the years companies have tried to improve ventilation systems and to encapsulate as many operations as possible. As a matter of fact some of the high activity concentrations reported above were reduced subsequently to 20 % and less by such improvements. As results indicate, various situations not complying with dose limits withstood all technical efforts to lower dust exposure. In these cases wearing of protection masks remains indispensable. Since these are not really popular among workers, compliance has to be checked by frequent inspections as well as by analysing mask filters. In any case, the actual working hours under each condition of inhalation exposure must be recorded and dose estimations are performed thereof. Yet it is hoped that once the excretion monitoring yields more reliable results, management and workers may be convinced of the effective doses and the benefits of minimising radiation hazards. Of course the most efficient and effective measure would be to abolish the use of thoriated material altogether. As a matter of fact, increasing the monitoring expenditure and insisting in the improvement of radiation safety has encouraged research and development of thorium substitutes such as cerium and lanthanum. Lacking legal grounds for banning the use of thoriated welding rods for instance, however, overloading the local producers would only increase cheap imports from countries with more lenient radiation control as can be observed already. Thus regulating workplaces with expected doses above 1 mSv/a as envisaged by the European Basic Safety Standards Directive, will hopefully help to overcome this problem.

Conclusion

Due to various inadequacies and shortcomings none of the various methods available to monitor internal doses was fund to be appropriate without restriction. Therefore so far no sufficiently significant correlation between the doses assessed by the different techniques could be ascertained and, for the time being, a combination of methods will be required. Nevertheless efforts to reduce doses by improving on-site installations will continue.

References

Lee D.; Skrable K.W.; French C.S.; Reevaluation of the committed dose equivalent from 232Th and its radioactive progeny. Health Phys. 72: 579-593; 1997.

Noßke D.; Modelle der ICRP für die interne Dosimetrie - Stand und Perspektiven; Proc.of the 30. Ann. Mtg. Of the German-Swiss and Austrian Rad. Pro. Assn.; Sep 28 - Oct 2 1998; Lindau, Germany; 253-259.

Eisenmenger A.; Brose J.; Scheler R.; Riedel W.; Inkorporationsüberwachung auf Thorium mittels 220Rn-Exhalationsanalyse; ibidem 177-181.

Sternad T.; Kratzel U.; Strahlenexposition beim berufsbedingten WIG-Schweißen mit thor. Wolframelektroden ; ibidem 27-32. Radiation protection in thorium processing industries - monitoring and minimising internal doses.