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Workplace exposure to ionising radiation in the United Kingdom from the use of naturally occurring radioactive materials

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WORKPLACE EXPOSURE TO IONISING RADIATION IN THE UNITED KINGDOM FROM THE USE OF NATURALLY OCCURRING RADIOACTIVE MATERIALS

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ABSTRACT

The National Radiological Protection Board (NRPB) has extensive experience of the radiological issues associated with the use of naturally occurring radioactive material (NORM) in a variety of industries in the United Kingdom. A selection of these is described and the radiological protection issues examined. Worker exposures due to inhalation and to gamma radiation are assessed for some of the industries discussed. Practical methods of dose restriction and workplace monitoring techniques are described.

INTRODUCTION

It has long been recognised that work with materials containing relatively low levels of naturally occurring radionuclides can give rise to significant exposure in the workforce[1]. In the UK, the Ionising Radiations Regulations 1985[2] and the associated Approved Code of Practice[3] specify activity concentrations of the naturally occurring radionuclides at which the material should be considered radioactive. The Approved Code of Practice states that "for substances which include the radionuclides thorium-232 or uranium-238, or any nuclides in their particular decay series, activity concentrations of the parent radionuclides exceeding 0.3 Bq g^{-1} or 1.0 Bq g^{-1} respectively for dusty operations, and 5 Bq g^{-1} or 9 Bq g^{-1} respectively for bulk storage" should be considered radioactive substances and hence subject to regulatory control. These values are based on earlier work by NRPB[1] on naturally-occurring radioactive materials (NORM) and reflect concern over the inhalation and external exposure pathways. This definition has brought many industrial processes under regulatory control and many Companies using these processes have used NRPB for specialist advice on radiation protection. NRPB has thus had the opportunity to investigate these processes in some detail. This paper reviews several processes that use naturally occurring radioactive materials, and estimates the radiation doses to workers, taking into account recent changes in metabolic models and revisions in dose coefficients.

INDUSTRIES WHERE THE POTENTIAL FOR EXPOSURE EXISTS

The problems associated with the build up of NORM in the offshore oil industry are well known[4]. However, many other industries also use feed materials containing naturally occurring radioactive materials. Typical activity concentrations of some of these materials are given in Table 1. From this it can be seen that activity concentrations can exceed the reference levels specified above. It is worth noting that in all of the applications considered, the presence of radioactivity is undesirable. Even where radionuclides are being deliberately added, as with some uses of thorium, it is because of the chemical and physical properties, not the radioactive properties.

A selection of the processes is described below to give representative examples of the radiological issues.

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Manufacture of zirconia

Baddeleyite is heat treated to alter its crystalline structure to form zirconia. After treatment the product is milled to achieve a range of small particle sizes and then packaged for sale. The process is relatively dusty and gamma ray dose rates are significant close to bulk material. Lead-210 and polonium-210 are volatile and are driven off during the heat treatment to deposit in the cooler parts of the effluent system. In particular, they occur at elevated activity concentrations in furnace flue dusts (see Table 1).

Zircon as a refractory

The use of zircon as a refractory material is a relatively common practice in steel foundries where a layer of zircon is used as the inner surface of a mould. Dusty conditions can exist in the preparation of the mould and during removal of surface deposits on the casting by abrasive techniques. Gamma dose rates can be significant near bulk material.

Zircon and zirconia in the manufacture of refractories.

Zircon and zirconia are used in the manufacture of refractory bricks and nozzles. Dusty conditions can exist during the mixing of the ingredients and gamma dose rates can be significant near to bulk storage. To date there is little evidence for the concentration of lead-210 and polonium-210 in furnace flue dusts.

Rare earths in glass polishing compounds

A heat treatment and particle size reduction process is used to condition the rare earth as a glass polishing compound. Dusty conditions can exist at the loading station and during product removal. Significant gamma ray dose rates exist close to bulk storage. When used as a polishing compound the radiological implications are generally not significant because of the small amounts being used.

Ilmenite and phosphate rock processing

Both these processes involve treatment with sulphuric acid. The insolubility of radium sulphate causes it to precipitate on the surfaces of tanks, pipes and filter frames where it can produce significant gamma dose rates. Contamination levels are relatively high and this is of radiological significance during descaling.

Rare earths in catalysts

A catalyst for the oil industry can be made by incorporating a rare earth mixture into a zeolite, but this is no longer carried out in the United Kingdom. The main radiological issue was the deposit of isotopes of radium in tanks and pipes where it presented a gamma dose rate problem and a contamination problem during descaling and decommissioning. As such, the problems are similar to those described above for ilmenite and phosphate rock processing.

Rare earths as an additive in special glasses

Rare earth concentrate is added as a minor component in certain special glasses. Dusty conditions can exist during loading and gamma dose rates are significant close to bulk storage.

Thoriated tungsten welding electrodes

The addition of thoria improves the striking performance of tungsten welding electrodes. In their manufacture the use of pure thoria is a major radiological protection problem due to inhalation and external radiation. In use, the electrodes need to be occasionally ground to restore a conical tip and dusty conditions can exist during this process.

Thorium magnesium alloys for aero engine components

Thorium is added to magnesium to produce a hardened, light alloy for use in aero engine components. The manufacture of the alloy could give rise to both inhalation and external radiation problems, but is no longer undertaken in the UK. There are, however, still some radiological implications in the aero engine refurbishment industry where thorium magnesium components may be subjected to abrasive processes creating dusty conditions.

Thorium fluoride as a lens coating

The process involves the vacuum coating of optical components to provide a non-reflective surface. The amounts of thorium in use are small but the widespread contamination produced in the vacuum chamber is problematic especially during cleaning procedures.

Offshore oil extraction

Sea water is used to displace the oil in the deposit. Dissolution and subsequent precipitation of the isotopes of radium cause scales with enhanced natural radioactivity to form in pipes, valves and storage tanks. Significant gamma dose rates can occur around such plant. When the plant is opened, both surface and air contamination can be a problem.

Offshore gas extraction

Radon may be present in the gas stream. As it decays the longer lived radionuclides, lead-210 and polonium-210 are formed and these deposit with sludges, and on surfaces and filters. Surface and air contamination are both potential problems. Gamma dose rates are negligible.

Titanium dioxide production via chlorination

Sources of titanium dioxide, such as rutile, are purified by means of a chlorination process in which titanium tetrachloride is distilled off. Dusty conditions can exist during loading and unloading of transport vehicles and during introduction of the material into the process. Gamma dose rates are usually not significant. Some concentration of radionuclides occurs in waste streams but these do not cause significant occupational exposure.

ASSESSMENT OF RADIATION EXPOSURE

NRPB has estimated the exposure of workers at a number of establishments using site-specific measurement data. For this paper, typical data from some of the processes described have been used to provide broad estimates of worker doses.

For external radiation exposure, Dixon[1] calculated that a dose rate of 2.5 μSv h^{-1} would be present close to bulk material containing either 5 kBq kg^{-1} of thorium-232 or 9 kBq kg^{-1} of uranium-238, in equilibrium with their daughters. The activity concentrations of several commonly used materials exceed these values, indicating that external exposure cannot be ignored from a radiological protection point of view. Dose rates close to bulk quantities of material have been calculated from the activity concentrations listed in Table 1. External doses have then been calculated using general assumptions regarding segregation and the occupancy factors in Table 2.

Doses from ingestion are generally trivial, even where no special precautions are taken, and only start to be important where high activity concentrations are present. Significant exposures from radon are theoretically possible but have been shown to be unimportant in the past[5]. Consequently, neither of these two exposure pathways is considered further.

Exposure due to inhalation of dust has been conventionally assumed to dominate and this assumption has been reinforced by previous work[1]. Many processes involve the handling of dry particulate materials and high dust levels are often encountered. The data used to assess intakes due to inhalation is shown in Table 2. The estimate of exposure following an intake depends on models of the metabolism of the radioactive material in the body, and these models have been considerably revised in recent years. The dose coefficients used have been taken from ICRP Publication 68[6] as revised by Publication 72[7] with respect to radium-226. It has been assumed that an activity mean aerodynamic diameter of 5 μm is appropriate. This is consistent with values found in industrial processes[8].

Estimated annual doses that might typically be received in selected industries are given in Table 3. It is clear from this that while exposures are well below dose limits they are not negligible. The estimates include the effect of respiratory protection in all but one case. Without such protection, worker doses would be substantially higher and in certain situations could approach or even exceed dose limits. Where respiratory protection is used the contribution from external radiation for some processes can be comparable with that from inhalation; the conventional wisdom that inhalation dominates in these situations may, therefore, have to be revised. The main reason for this is the revised dose coefficients that have reduced inhalation doses by a factor of between two and ten, depending upon the materials being used[9].

WORKPLACE MONITORING AND THE INTERPRETATION OF RESULTS

Analysis of materials

The recommended starting point of all radiological assessments of work with NORM is determining which radionuclides are present and at what activity concentrations. In the first case this information may be used to estimate the hazards that might exist in, say, dusty processes, and to determine the regulatory position. Secondly, this information is usually required to interpret workplace monitoring results, as will be discussed later.

Methods for the analysis of the radioactive content of NORM are essentially of four kinds, namely gamma-spectrometry, alpha spectrometry, neutron activation and other methods (eg, chemical, X-ray fluorescence, mass spectrometry and atomic absorption).

Gamma spectrometry is a relatively simple and inexpensive technique but it only provides direct information on the activity concentrations of the gamma emitting radionuclides. As such, there is no direct measurement of radiologically important isotopes of uranium and thorium or the radionuclides, lead-210 and polonium-210. Making assumptions about the state of the equilibrium within the decay series is often necessary. Evidence for this may be present in the gamma spectrometry results and the history of the material. Scales and other deposits are a case in point. Surface deposits that have resulted from the precipitation of radium salts are likely to consist of radium-226 and radium-228 in equilibrium with their daughters but with the parent radionuclides thorium-232 and uranium-238 essentially absent. Deposits resulting from the transfer of radon are likely to consist of lead-210 and polonium-210 but not necessarily in equilibrium.

Chemical analyses and neutron activation techniques provide a very valuable adjunct to gamma spectrometry when disequilibrium is suspected. Both techniques provide results for the radionuclides that are gravimetrically the most abundant. They can provide the necessary information for isotopes of thorium and uranium and a combination of such methods and gamma spectrometry is often required. In addition, the determination of polonium-210 (by alpha spectrometry) may need to be considered, especially where radon daughters have accumulated or high temperature processes have occurred.

External radiation

Monitoring is required for external gamma dose rates to designate areas and estimate individual doses. This is readily achieved by means of portable radiation monitoring equipment using simple techniques. Individual dosimetry has traditionally been considered unnecessary although the estimated doses in Table 3 suggest that this needs to be reviewed for certain processes.

Internal radiation

Surface contamination

The type of measurement undertaken, and indeed the purpose of monitoring, depend upon the activity concentration of the material. At high activity concentrations, for example thorium compounds and some radium scales and polonium furnace dusts, the use of surface contamination monitors is necessary to check that adequate contamination control is being maintained and to designate areas. Often, however, there can be difficulties with self-absorption (for beta and especially alpha monitoring) and ambient dose rates (for beta monitoring). In addition, the interpretation of results requires a knowledge of the state of equilibrium in the two major decay series. This may well be specific for the material that is being used and may even vary with its age. In such cases, removing a sample of the material from the surface to analyse the actual radionuclide content may be more reliable.

For low activity concentration materials, such as ores, the emphasis changes from restricting the spread of surface contamination (which is often not practicable) to minimising the resuspension of dust. In fact, significant levels of surface contamination are usually visible as relatively thick deposits of material. With such materials, a visual inspection of the workplace is likely to be adequate for assessment and control, provided the areas are otherwise relatively clean. If quantitative assessments of surface contamination are needed this can be achieved by gravimetric sampling of measured areas. The surface concentration of individual radionuclides can then be assessed by reference back to the activity concentrations of these radionuclides in the bulk material.

Air sampling

In all the processes considered, air sampling is necessary, at least on a trial basis, to establish the ~~likely inhalation doses and to designate areas. Generally, this is achieved by personal air sampling.~~
Once a sample has been obtained, the air concentration can be determined by:

(i) counting of emitted radiations;
(ii) other analytical methods, such as X-ray diffraction or mass spectrometry of the sample, in which the thorium or uranium content may be measured or inferred; or
(ii) gravimetric assessment.

The first technique usually involves counting total alpha emissions. The air concentrations of specific radionuclides are then estimated from a knowledge of the radionuclide composition of the materials being used. Most employers do not, however, own the required counting equipment and the other methods listed above may often be more convenient. Again, a knowledge of the radionuclide composition of the material is necessary to interpret the results.

The last technique is usually the most convenient for the employer who already undertakes dust measurements. If the activity concentrations of individual radionuclides are known (ie by suitable analysis of the materials), it is possible to derive specific reference levels in terms that are already familiar to occupational hygienists (eg in units of mg m^{-3}). Caution needs to be exercised however in two respects. First, the airborne dust may contain materials other than the radioactive material; reliance on dust level measurements will then overestimate the potential radiation exposure. Second, some materials have been known to have an increasing activity concentration with decreasing particle size. It is possible, therefore, that dust in the atmosphere will have a higher activity concentration than the bulk material and so radiation exposures might be underestimated.

DOSE RESTRICTION IN PRACTICE

Inhalation

Many industrial processes involving NORM have been in operation for many years, and inevitably involve dusty operations. Ideally, dose restriction should be achieved primarily by engineering controls that prevent airborne dust in the first place. However, while engineering controls can be readily introduced in the planning stage of a new process or facility, their application can be more problematic in old, established processes. Also, the practicality and effectiveness of such controls depend upon the scale of the operation. Small scale processes, such as grinding thoriated welding rods or the use of thorium compounds in lens coating, can be effectively controlled by using enclosures and localised extraction. In large scale processes, such as the bulk processing of ores, complete containment is not practical. In these situations, protection measures consist of a combination of plant modifications, ventilation, working procedures and, where necessary, respiratory protection. The plant modifications are essentially the same as those used for nuisance dust control and typically fall into two categories. The first is improved methods of transferring materials through the plant, for example, the use of enclosed screw feeds rather than batch-transfers using open hoppers. The second category is improved containment and dust extraction on existing plant. Even with such modifications, it is still common for personal respiratory protection to be necessary in certain circumstances. The most common type is the disposable half-face dust mask and these tend to be worn for specific tasks rather than continuously. For example, respirators will be worn when entering certain parts of the plant (eg mill enclosures and bag houses) or when particularly dusty jobs are undertaken (emptying or filling bags, cleaning plant, etc).

In all processes, good working procedures are necessary to ensure that proper dust control is maintained. In many processes, the materials handled have historically been regarded as "non-toxic". In these cases, training to appreciate the hazards is an important step.

External radiation

Both gamma radiation and beta radiation could in principle contribute to external radiation exposure. In practice, however, only the gamma radiation is important. Significant gamma dose rates exist close to higher activity concentration material such as thorium oxide, although the amounts of material in use are usually small and dose rates reduce quickly with distance. Thus, avoidance of exposure is a simple matter of distance and time restriction. Additional shielding may also be practicable in storage situations but may not be strictly necessary.

Significant gamma dose rates may also exist close to widespread deposits of medium specific activity material, such as scales and sludges, and bulk amounts of low activity concentration material such as ores. For scales and sludge, where the dose rates warrant it, simple barriers can be effective in restricting close access and thereby reducing exposure. For ores, the large quantities involved, and factors such as the need to transport feed materials into the process, make the provision of additional shielding costly or difficult. There is also a much slower reduction of dose rates with distance than with small amounts of material. Avoidance is usually the most practical option and it makes sense to store bulk material away from routinely occupied areas. Simple measures such as signs or floor markings instructing employees not to loiter are usually effective.

CONCLUSIONS

Some of the industrial uses of materials containing NORM undertaken in the United Kingdom have been described. The radiation exposure pathways associated with these processes have been discussed and the typical radiation doses that workers receive have been estimated. In at least some of these processes, radiation exposures from inhalation and external radiation are sufficient to warrant further action. The processes and specific site situations are many and varied and thus case specific assessments of the radiological hazards are necessary to ensure appropriate protection is instituted. For any such assessment, it is suggested that the radionuclide composition of process materials needs to be known to the assess the likely radiation hazards, and to interpret workplace monitoring results. Finally, practical methods which have been used to restrict radiation exposures in the UK are described.

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Table 1 Typical activity concentrations of material in use (kBq kg^{-1})

Material	Thorium-232	Radium-228	Uranium-238	Radium-226	Lead 210	Polonium-210
Zirconia/baddeleyite	0.3	6	7	7	7	7
Zirconia flue dusts	0.5	8	3	3	200	600
Zircon	0.6	0.6	3	3	3	3
Rare earth	6.0	6.0	1	1	1	1
Thoria	3600	1800	0	0	0	0
Thorium magnesium alloy	160	80	0	0	0	0
Thoriated tungsten	160	80	0	0	0	0
Thorium fluoride	3600	1900	0	0	0	0
Oil extraction scale	0	5	0	40	0	0
Gas extraction deposits	0	0	0	0	50	100
Phosphoric acid scale	0	20	0	100	100	100
Ilmenite scale	0	20	0	100	100	100

Note: The activity concentrations in thoria, thorium magnesium, thoriated tungsten and thorium fluoride are dependent on the time lapse since the pure thorium compound was prepared.

Table 2 Factors used in assessing typical exposures

Process	Dust level (mg m^{-3})	Hours per day	Days per year	Average respirator protection factor
Production of ZrO_2	5	7	200	1
Fume	10	2	50	5
Zircon	5	1	10	5
Polishing compound	5	3	200	10
Th/Mg alloys	5	2	50	10
Thorium fluoride	5	1	20	10
Oil extraction scale	5	1	10	10
Gas extraction	5	1	20	10
Process scales	5	1	20	10

Table 3 Summary of estimated typical doses

	Estimated annual effective dose (mSv)		
Process	External radiation	Inhalation	Total
Production of ZrO_2	0.5	2.3	2.8
Fume	0.4	0.4	0.8
Zircon	0.0	0.0	0.0
Polishing compound	0.4	0.1	0.5
Th/Mg alloys	2.5	6.6	9.1
Thorium fluoride	2.0	1.3	3.7
Oil extraction scale	0.1	0.0	0.1
Process scales	0.4	0.0	0.4