WASTE CONTAINING ENHANCED CONCENTRATION OF NATURALLY OCCURRING RADIONUCLIDES

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SUMMARY: Since radiation risk is usually considered to be related to nuclear energy or atomic bomb, majority of researches on radiation protection have focused on artificial radionuclides and radioactive waste. Far less attention has been paid to radiation risk caused by exposure to ionizing radiation originating from naturally occurring radioactive materials (NORM) despite the fact that their presence touches many aspects of our life, starting with occupational risk, through some "contaminated" goods, leisure activities including spa visits and ending with a huge amount of bulk waste often dumped in our vicinity. Each particular occurrence of natural radioactivity presents a unique scenario of exposure – usually different from those caused by artificial radionuclides. In the article the cases of tailings containing enhanced natural radioactivity are identified. Proposed rules of such materials classification and method of unification with other type of waste are described.

1. INTRODUCTION

Despite of the history of artificial radionuclides (^{137}Cs, ^{90}Sr etc.), their influence on the environment is limited to only few decades and an overwhelming majority of present-day radioecological investigations is connected with study of redistribution in environment and biological action of this group of radionuclides. Far less attention was paid to radiation risk to people and environment caused by expose to enhanced ionizing radiation originating from naturally occurring radioactive materials. But after the occurrence of natural radioactivity had been thoroughly studied, it became clear that such phenomena are present very frequently in human environment. Many processes in non-nuclear industry create a situation when the concentration of naturally occurring radioactive material is additionally enhanced. Such situation usually exists in industrial processes where a significant mass reduction of raw materials occurs, typically with changes of their chemical composition or state of aggregation which might further influence their properties. As a matter of course, these industries of concern are not aimed at the production of natural radionuclides or the deliberate use of radiation. Therefore radioactive isotopes are usually accumulated in the waste. Such alterations to the natural state result in an increment of radiation risk to people as well as to the environment in case of disposal of such

waste. Each particular occurrence of natural radioactivity presents a unique scenario of exposure – usually different from those caused by artificial radionuclides.
Usually the amount of such type of waste materials can be up to hundreds of thousands of cubic meters or tonnes; and material, which is often associated with other pollutants as heavy metals or hydrocarbons has been dumped directly into the environment. Therefore the application of routine rules used for assessment of risk caused by artificial radioactivity (i.e. radioactive waste) and practices[1] can lead to the complete misunderstandings.

The importance of radiation risk caused by natural radioactivity firstly have been underlined in the Council Directive 96/29EURATON laying down basic safety standards for the protection of the health of workers and the general public against the danger arising from ionizing radiation. Namely, in the paragraph 40 of this Decree, it is clearly stated that common rules of radiation protection must be applied in following cases:

- work activities involving operations with, and storage of, materials, not usually regarded as radioactive but which contain naturally occurring radionuclides, causing a significant increase in the exposure of workers, and, where appropriate, members of the public.
- work activities which lead to the production of residues not usually regarded as radioactive but which contain naturally occurring radionuclides, causing a significant increase in the exposure of members of the public, and, where appropriate workers.

2. SYSTEM OF NATURAL RADIOACTIVITY OCCURRENCE CLASSIFICATION

After the EC Directive 96/29 has been published the problem of risk caused by natural radioactivity was widely discussed. Some authors have followed terminology applied by Gesell and Pritchard [1975] [i.e. Kathren, 1998, Righi et all 2000] but a lot of different abbreviations have emerged to describe these phenomena. For example Baxter [1996] applied TERM (technologically enhanced radioactive material), Vandenhove [2000] used “materials containing natural radionuclides in enhanced NORs”. In Canada two names: NORM- contaminated and Naturally Occurring Nuclear Substances have been applied to distinguish non-nuclear industry from uranium ore extraction and processing (Ministry of Health, Canada, 2000). A. Martin in the first report dealing with European industries working with natural radioactivity used simple descriptive name “materials containing natural radionuclides in enhanced concentrations” [Martin et al.,1997]. IAEA [2003] report has distinguished two situations when natural radioactivity in non-nuclear industry can cause significant increment of radiation risk: NORM (naturally occurring radioactive materials) and technologically enhanced NORM. Such approach was followed in many articles presented at the periodic conference NORM IV held in Poland [IAEA 2005]. All mentioned above, abbreviations are used by authors interchangeably to describe situation when presence of the natural radioactivity causes a non-negligible radiation risk. But even after a preliminary analysis of branches of minerals processing industry it is clear that such enhanced risk can be present in two different ways and the best way to describe them are acronyms NORM and technologically enhanced NORM (TENORM).

The term NORM (Naturally Occurring Radioactive Materials) should be used, accordingly to the definition, only for cases, when radiation hazard is due to the presence of materials with elevated concentration of natural radionuclides, significantly above average level of radioactivity, albeit not related to or caused by any type of human’s activity. It has to be pointed

[1] Activity focused on an use of radioactive materials, ionizing radiation or fissile materials according to the definition given by IAEA

out, that NORMs are taken into account in the radiation hazard assessment scenarios only in cases when they appear in the natural or work environment due to industrial activity. Otherwise these materials are treated as sources of the natural background and are not taken into consideration as an enhanced radiation risk.

The acronym TENORM(s) means Technologically Enhanced Naturally Occurring Radioactive Materials. This term is used for the description of any raw material, product or waste, in which concentrations of the natural radionuclides have been altered (enhanced) as a result of technological processes to the levels causing significant increase of the radiation hazard above natural background. It is irrelevant if this enhancement is intentional or not. In many cases NORMs are used as a raw material for a process where TENORMs are created as products or by-products. On the other hand, it's possible to generate TENORMs in processes, where no NORMs have been used as raw materials. An example of such processes is coal combustion for power generation. Hard coal is well known as a material with relatively low concentration of natural radionuclides, and cannot be treated as NORM at all. The combustion leads to the very big reduction of the initial fuel mass. Owing mainly to the elimination of organic component of hard coal, there is approximately one order of magnitude enhancement of the radioactivity concentration from fuel to ash during the combustion process.

One should remember that raw materials and substances used in the nuclear industry either for civil or for military purposes are not classified as TENORM. In addition, in the current terminology applied in radiation protection such activities are classified as "practices" in comparison with those dealing with NORM or TENORM that are named "work activities".

3. RADIATION RISK SCENARIOS

According to definitions mentioned above, processes leading to the increase of the radiation hazard due to natural radioactivity can be divided into two groups.

The first one includes exploitation, transfer or disposal to the natural environment of raw materials or waste without any changes of their properties. Such processes are mainly performed in the industrial branches, relating to exploitation of mineral resources with elevated concentrations of natural radionuclides – phosphates, tin, titanium, niobium and rare earth metal ores. The waste rock produced in the exploitation processes is in form of natural materials, with the same chemical composition and physical properties. The enhancement of radiation hazard is usually a result of the direct exposure to its radiation and may be due to increase of the amount of waste in the installation or its release to the natural environment. Also, different exploitation technologies applied in underground mining or drilling may lead to unintentional release of waters or gases with elevated levels of natural radioactivity. The presence of NORM is in this case may create significant radiation risk. Following waste materials can be listed as mentioned above substances:

- Waste rocks from mining exploitation and raw materials enrichment, together with possible contamination resulting from leaching of natural radionuclides;
- Brines, released from oil and gas rigs or hard coal mines;
- Radon and its progeny in caves, underground mines, in tunneling and in natural gas;
- Dust from systems of air cleaning.

The second group consists of processes leading to significant mass reduction, typically with changes of chemical composition or state of aggregation which might further influence behaviour of radioactive elements in the environment. Such processes are characteristic for industries processing raw materials. Mass reduction in these processes may cause concentration of all impurities in produced waste materials, together with natural radioactivity. Wastes with concentration of natural radionuclides in order of one or two magnitudes higher than in the raw materials are often created as the result of such processes. Besides fossil fuels combustion, all parts of the processes of metal ores treatment (metallurgy) as well as particular stages of certain inorganic chemical technologies can be included to this category. In this case to create radiation risk it is not necessary to process NORM materials and these raw materials in practice are often contain less natural radioactivity than usually taken as average for Earth's crust.

Properties of the waste materials resulting from the second group of technologies are related to the level of accumulation of the natural radionuclides due to the mass reduction within the process. Following waste materials can be included into this category:

- Waste from fossil fuel combustion;
- Waste materials from chemical technologies (mainly production of phosphoric acid and titanium dioxide pigment);
- Waste from metallurgical industry (iron ore processing and steel production);
- Waste products from non-ferrous ore processing – mainly tin, copper and niobium;
- Waste from rare earth metals industry.
- Deposits in underground galleries and settling ponds, scales in the dewatering systems in well and underground mining industry;

Besides the occupational risk and possible risk to the members of the public inhabiting neighboring areas NORM/TENORM type waste impacts upon the environment where it has been dumped. After the placement into the environment this type of waste may set some additional processes in motion, leading to the selective transfer and accumulation of particular radionuclides and disequilibrium in chains of natural radioisotopes.

4. REGULATION INCONCLUSIVENESS

In each EC member state the national Atomic Acts and subsequent enactments establish a uniform legal framework for regulatory control of activities subjected to radiation risk. Such activities are traditionally considered to be directly related to the intentional use of ionizing radiation, radioactive sources or fissile materials. These activities are carried out under specified conditions and monitoring of radiation risk is obvious. The radiological protection monitoring parameters are source activity, kind of radiation, dose rate, etc. Regulatory control steps in just when the radiation risk exceeds the allowed limits. Hence, in such situations one can distinguish two levels of radiation risk monitoring. The first can be called "physical control", that is carried in each of the above situations and its main objective is to determine when the level of the risk is close to the limits. The second one, regulatory control, is carried out above the specific levels of radiation doses and usually legitimizes the use of radiation, but it also forces a user to keep the radiation risk below the level allowed by relevant law.

Such approach is well-founded and effective only in the case when the physical control of radiation risk is assured or at least attempted. Otherwise a vicious circle can be a result: there is no information about radiation risk in the absence of physical control, so that, there is no reason to introduce the regulatory control. As there is no regulatory control, there is no driving force to start any activity focused on radiation risk monitoring. Finally, even severe radiation risk can be denied.

In case of radiation risk caused by enhanced natural radioactivity the necessity of physical control is not obvious. Natural radioactivity is a primordial property of the surrounding matter. Natural radionuclides are present in almost all substances that we deal with. Therefore, the generic question is: when is their presence significant from a radiation protection point of view? The answer seems to be very simple: when the derived radiation risk exceeds acceptable level, it means, when the necessity of regulatory control appears. But to get such answer is not so simple. In case of exposure to the risk caused by natural radioactivity there is usually no physical control in absence of detailed regulations. This results in the gap of knowledge about the real radiation risk. So, no one knows whether allowed limits of risk have been exceeded or not, hence the problem of regulatory control does not exist. This implies that there are no risk evaluation and no needs for physical control. In this way quite often even serious radiation risk can be out of system of control of radiation protection.

On the other hand, the problem of natural radioactivity is completely excluded from regulations dealing with the environmental protection in general. Finally, hazard caused by radiation originating from naturally radioactive materials is rarely taken into consideration when the treatment of industrial waste is planed.

5. PROBLEM SOLUTION

To solve the mentioned problem one would enforce the physical control of the radiation risk in each industry dealing with exploration or processing of natural resources in general. But such approach would be a substantial overuse and, due to actual level of knowledge in this matter, is not justified.

Solution applied in many countries and also recommended by IAEA is to identify the industries where presence of natural radioactivity can cause significant radiation risk. In comparison with mentioned EC directive, where only examples as underground mining and caves were listed, so called “positive list” currently contains eleven branches of the industry. Existence of this list results in the obligatory monitoring of radiation risk in the companies concerned. Such approach makes one actually apply the requirements and regulation that have been developed in order to control risk caused by artificial radioactivity, including exemption or clearance levels expressed as mass or volume activity concentration. But the direct application of these rules and recommendation to TENORM management lead to many discrepancies or even paradoxes. For example, usually amount of TENORM type waste is so big that taking into account total activity concentration criterion almost always results in an outcome that it should be treated as radioactive waste. For the same reason there is no possibility to fulfil all requirements related to radioactive waste.

On the other hand the problem of NORM/TENORM is completely pushed away from acts ruling the treatment of industrial waste and environment preservation or health protection. Actually ionising radiation due to its carcinogenic and mutagenic properties is mentioned in these acts but typically only in the form of a reference to regulations dealing with radiation protection and the circle is closing again.

The solution based on introduction of the “positive list” into a national regulation, but it is still a substantial generalization and does not provide the end user with help on how to cope with the problem. But if one considers thoroughly TERNORM occurring as waste, the possibility to define in detail the circumstances of radiation risk creation on case-by-case basis is available. Namely, European Waste Catalogue (EWC) seems to be a good basis to collect all information about a particular waste, including properties important form radiation protection point of view. Such idea is additionally supported by the fact that TENORM type waste looks like “common” industrial waste rather than nuclear or radioactive one. Also the consequences of TENORM occurrence in the waste are often amplified by the simultaneous presence of other pollutants.

The European Waste Catalogue was produced following the Commission's decision[2] and it is a fundamental part of a safe waste disposal. It classifies both hazardous and non-hazardous waste produced pursuant to European Council *Directive 75/442/EEC of 15 July 1975 on waste* and categorizes them according to what they are and how they were produced. The catalogue defines standardized nomenclatures and monitoring levels of the various waste types. The EWC codes are valid throughout Europe and contain just about any waste conceivable. The EWC defines the basic necessity of monitoring certain wastes but the monitoring level can be adapted to one's special needs because of requirements imposed by authorities, customers or internal regulations.

The preliminary analysis of EWC in relation to literature available TENORM data showed that waste accounted as *wastes from thermal processes* (group 10) are the most numerous category well known as containing enhanced concentration of natural radionuclides. It is caused mainly by concentration of radioactive isotopes during such processes as combustion, melting, evaporation etc. As one should have expected, also the groups 01 and 06, *Waste resulting from exploration, mining, quarrying, physical and chemical treatment of minerals* and *waste from petroleum refining, natural gas purification and pyrolytic treatment of coal,* respectively contain a multitude of different waste materials where high concentrations of natural radioactivity is a primordial property of these waste, or have been obtained due the technological process, or resulted from unwanted side effects. In the light of the common radiation protection requirements, wastes listed in these groups are the most important from point of view of the expected radionuclide activity concentrations, as well as their total amount usually produced.

Among individual waste already classified in EWC more than 240 are at least suspected as being TENORM. In spite of the EWC containing a very large number of different types of waste sometimes it seems worth distinguishing a new category of individual waste, taking into consideration the concentrations of natural radionuclides. For example, sludge settled at the bottom of ponds in coal mining industry may have so high activity concentration of radium that it deserves to be separated from other waste classified as subgroup *01 01 wastes from mineral excavation.*

6. CONCLUSION

In the light of other European regulations that very often contain a lot of details concerning subjects that in comparison with radiation risks appear not to be so serious and with many negative consequences, the EC Directive 96/29/EURATOM appears not to include as many details as it is necessary. The results is that the problems of NORM/TENORM are mostly out of any regulation and non-nuclear industry of concern was until now not aware of the problems connected with natural radioactivity and may also expect some negative consequences in case of implementing radiation protection measures. Therefore many companies do not provide radioactivity data as long as no precise regulation exists. Finally, in spite of the number of reports issued recently and describing effects of TENORM and NORM occurrence in industry and environment this kind of risk is not sufficiently appreciated. There is a great need to provide mineral industry operators with well-founded information about real risks caused by enhanced natural radioactivity.

[2] The EWC was originally established by Commission Decision 94/3/EC. It was replaced by 2000/532/EC and amended by Decisions 2001/118/EC, 2001/119/EC and 2001/216/EC. The full EWC is available from the European Commission's web-site at:
http/europa.eu.int/eur-lex/en/lif/reg/en_register_15103030.html

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