

ASSESSMENT OF GAS PRODUCING RADIOACTIVE WASTE DISPOSAL

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In near-surface repositories for radioactive waste, significant quantities of gases may be generated as a result of microbial degradation and corrosion. The potential impact of gas generation, accumulation and migration on the long-term safety of a repository, should therefore be assessed properly. We present here safety assessment results of gas producing radioactive waste disposal as well as gas generation calculations.

Key words: radioactive waste, disposal, gas generation, biodegradation, corrosion

1 INTRODUCTION

Low and intermediate level radioactive waste is produced mainly by the nuclear sector in Slovakia. It contains materials contaminated by radioactive substances and is commonly disposed of by placing in near surface repository (RU RAO Mochovce). Some of the radioactive waste (RAW), particularly non-segregated pre-compacted solid waste from NPP A-1 contains cellulose and other degrading and gas generating materials. This radioactive waste is intended to be disposed of without pre-treatment by incineration to destroy the organic components, because sorting of this kind of waste is complicated, expensive and hazardous. However, radionuclides in such a waste may migrate through engineered barriers and easily reach the wider environment. Safety assessment therefore requires a detailed understanding of gas generation and transport within the disposal system. Concerning the issue mentioned above, safety assessment was aimed to quantify accurately the risk to human health.

2 GAS GENERATION MECHANISMS

The following three primary gas generation mechanisms have to be taken into account:

- Metal corrosion (iron and aluminum);
- Microbial degradation (cellulose, wood, cloths, rubber); and
- Radiolysis (bitumen matrix and water).

It has been proved, that the most important mechanisms are firstly corrosion (specifically anoxic corrosion) of steels and other iron alloys, as well as aluminum alloys; and secondly microbial degradation (specifically anaerobic microbial degradation) of cellulose, and perhaps rubber, cloths and plastic. A number of studies indicate that gas generation rates from radiolysis are not as significant as compared to the rates expected from anoxic corrosion and anaerobic microbial degradation [2]. Gas generation

is synergistically dependent on the conditions within the repository, with particular emphasis on the residual water content in waste at the time of emplacement and the water inflow from the surrounding formation. A number of laboratory gas generation studies have shown that the water content in the repository has a direct effect on the gas generation rates [3].

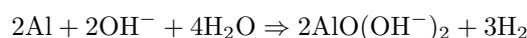
2.1 Corrosion

As mentioned earlier, the corrosion of metals in the repository may produce significant amount of gas. The quantity of gas production is strongly dependent on two major factors: (1) the amount of water present in the waste at the time of emplacement and/or the amount of water that enters the repository after closure and (2) the quantity of corrodible metals present in the waste.

There are two corrosion mechanisms that can occur in the disposal facility: oxidic corrosion and anoxic corrosion. Oxidic corrosion of the corrodible metals in the waste inventory will consume the oxygen in the disposal containers. After the depletion of oxygen initially present in the repository, anoxic corrosion of these materials will dominate. This will produce hydrogen and consume water at rates that will depend on the amount of water present [2]. Chemical reactions of the iron metals corrosion process are as follows [4]:



Aluminium is not thermodynamically stable in water, but has a protective oxide layer. However, in alkaline environments this oxide layer will dissolve and a rapid corrosion of aluminium yielding hydrogen can take place according to the general reaction:



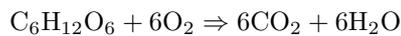
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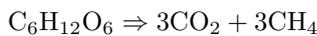
In alkaline environments aluminium alloy corrodes rapidly and the corrosion rate can be in a range of 10^{-3} to 10^{-2} m per year, equal to a hydrogen evolution of 0.15 to 1.5 kmol per square meter and year. Complete corrosion of 1 kg of aluminium corresponds to a hydrogen evolution of about 0.06 kmol [6].

2.2 Microbial Degradation

The principal source of microbiological gas production in ILW and LLW is expected to be the degradation of cellulosic materials such as paper, wood, and tissues. Microbial degradation can take place under both aerobic and anaerobic conditions. The general reaction of aerobic cellulose degradation is the following:



After the depletion of oxygen in the repository, anaerobic microbiological degradation will begin. From the standpoint of the gas and water contents of the repository, anaerobic microbial degradation will be much more intensive than the aerobic one because there will be much more nitrate and sulphate ions and carbon dioxide present than oxygen [2]. Assuming moisty conditions, anaerobic microbial degradation in the disposal rooms is given by the following reaction:



The final end products in an anaerobic environment may be carbon dioxide, hydrogen and methane. Existing rates of microbial degradation are often given as moles of total gas produced annually per kg, which is sufficient for substances of large surface areas. On the other hand, rates given as moles of gas produced per square meter are more valid for substances having low surface to volume ratios, such as bitumen.

2.3 Radiolysis

Radiolysis of water in the waste and in the repository will consume water and produce hydrogen and perhaps limited amount of oxygen. A variety of gases can also be produced by the alpha radiolysis of cellulose, rubber, and plastic in the waste. It can be concluded that alpha radiolysis of water will produce hydrogen and oxygen at rates much lower than the expected gas production rates for anoxic corrosion and anaerobic microbial degradation [1]. Additional evaluation proved that radiolysis of cellulose, bitumen and plastic will be minimal from the standpoint of long-term gas production in the repository [3], that is why this process was neglected.

3 RAW DESCRIPTION

Radioactive waste consists of aluminium scrap, steel plates, paper and tissues. Weight of RAW is 1500 kg, contamination by ^{14}C is about 2.5×10^6 Bq per waste package. Material composition of the RAW is given in the Table 1.

Table 1. Material composition of the RAW

Type of material	Fraction (%)
Plastic	10
Tissues	10
Concrete	10
Metals (90% Fe)	60
Paper	10

Table 2. Gas generation results

Type of RAW	G (m^3y^{-1})	V_{max} (m^3)	Max. amount
Steel – drums	0.345	517.50	20 pcs
Steel – RAW	0.481	962.00	900 kg
Aluminium	4.087	81.74	45 kg
Cellulose	0.219	248.79	300 kg
TOTAL	5.132	1810.03	

The final waste package is assumed to be fibre-reinforced concrete (FRC) container. It is of cube shape with outer dimension $1.7 \times 1.7 \times 1.7$ m and consists of a container body, a cover and two caps. The FRC is intended for storage and disposal of low and intermediate radioactive waste. High quality of container wall material was defined and confirmed by French Radioactive Waste Management Agency ANDRA. Usage of this type of container was approved by Nuclear Regulatory Authority of Slovak Republic.

4 CALCULATION RESULTS

4.1 Gas Generation Calculation

It is assumed in the presented mathematical model that conditions for microbiological processes and corrosion are optimal (very conservative approach). Gas generation due to corrosion of metals is given by:

$$G = Sr\rho l/M_v X V_0$$

- S total outer surface (m^2)
- r corrosion rate (mm y^{-1})
- ρ volume mass (kg m^{-3})
- M_v mass of the metal per kmol (kg/kmol)
- X stoichiometric coefficient (kmol/kmol)
- V_0 molar volume of ideal gas (m^3/kmol)

The rate of gas production due to microbial degradation was calculated as follows:

$$G = M_{\text{cel}} R_{\text{bio}}$$

- M_{cel} total amount of cellulose (kg)
- R_{bio} specific rate of microbial degradation ($\text{m}^3\text{kg}^{-1}\text{s}^{-1}$)

The calculated annual gas production, total production of gas and maximal amount of biodegrading radioactive waste in FRC container are given in the Tab. 2.

4.2 Dose Assessment

The gases of primary interest in post-closure radiological safety assessment are $^{14}\text{CH}_4$ and $^{14}\text{CO}_2$. These gases result in radiation exposure of inhabitants living nearby repository. The effective dose of this critical group can be expressed as:

$$D = D_{\text{inh}} + D_{\text{ing}} + D_{\text{ext}}$$

As only gaseous radioactive products are taken into account this time, the total dose is given mainly by inhalation as follows [5].

$$D_{\text{inh}} = C_{\text{air}} T_{\text{out}} B_{\text{air}} DC_{\text{inh}}$$

$$D_{\text{inh}} = 2.952 \times 10^{-2} \text{ Bq m}^{-3} \times 8766 \text{ h y}^{-1} \times 1.0 \text{ m}^3 \text{ h}^{-1} \times 5.8 \times 10^{-9} \text{ Sv Bq}^{-1} = 1.5 \times 10^{-6} \text{ Sv y}^{-1}$$

C_{air} contamination of the air (Bq m^{-3})
 T_{out} occupancy nearby repository (h y^{-1})
 B_{air} average breathing rate of a human ($\text{m}^3 \text{ h}^{-1}$)
 DC_{inh} dose coefficient for inhalation (Sv Bq^{-1})

The associated air concentration of a radionuclide can be approximated by:

$$C_{\text{air}} = R_{\text{gas}}/V_{\text{air}}$$

R_{gas} release rate of radioactivity associated with the gas (Bq y^{-1})
 V_{air} volume of the air into which the activity released per year is diluted ($\text{m}^3 \text{ y}^{-1}$)

The release rate in gas R_{gas} was calculated as follows:

$$R_{\text{gas}} = A_r f_{\text{gas}}/\tau_{\text{gas}}$$

A_r residual activity (Bq)
 f_{gas} fraction of the activity associated with the gas (-)
 τ_{gas} average timescale of gas generation (y)

The volume of the air above the repository site, where the contaminated gas is effectively diluted is given by:

$$V_{\text{air}} = Wuht$$

W width of the repository perpendicular to the wind direction (m)
 u average wind speed (m s^{-1})
 h height for vertical mixing (m)
 t the number of seconds in a year (s y^{-1})

5 CONCLUSION

This assessment was focused on the production of hazardous gases by means of biodegradation and metal corrosion. Calculated maximal annual individual dose for a member of a critical group is $1.5 \times 10^{-6} \text{ Sv y}^{-1}$. This figure represents just 1.5% of the dose limit for evolution scenario of Mochovce repository.

Safety assessment results showed that non-segregated pre-compacted solid waste containing cellulose, aluminium, iron and other degrading and gas generating materials may be safely disposed using FRC containers.

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