Journal of ELECTRICAL ENGINEERING, VOL. 57, NO. 4, 2006, 235–237

COMMUNICATIONS

ASSESSMENT OF GAS PRODUCING RADIOACTIVE WASTE DISPOSAL

Jozef Prítrský [*] — **Igor Matejovič** [*]
František Ondra [*] — **Vladimír Nečas** [**]

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.

K e y w o r d s: 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: oxic corrosion and anoxic corrosion. Oxic 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]:

aerobic corrosion	$4\mathrm{Fe} + 3\mathrm{O}_2 \Rightarrow 2\mathrm{Fe}_2\mathrm{O}_3$
oxide-film reduction	$4\mathrm{Fe}_2\mathrm{O}_3 + \mathrm{Fe} \Rightarrow 3\mathrm{Fe}_3\ O_4$
anaerobic corrosion	$3\mathrm{Fe} + 4\mathrm{H}_2O \Rightarrow \mathrm{Fe}_3\mathrm{O}_4 + 4\mathrm{H}_2$

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:

$$2\mathrm{Al} + 2\mathrm{OH}^- + 4\mathrm{H}_2\mathrm{O} \Rightarrow 2\mathrm{AlO}(\mathrm{OH}^-)_2 + 3\mathrm{H}_2$$

[*] DECOM Slovakia, Ltd., J. Bottu 2, 917 01 Trnava, Slovakia; E-mail: pritrsky@decom.sk, matejovic@decom.sk, ondra@decom.sk
[**] Slovak University of Technology, Faculty of Electrical Engineering and Information Technology, Department of Nuclear Physics and Technology, Ilkovičova 3, 812 19 Bratislava; E-mail: vladimir.necas@stuba.sk

ISSN 1335-3632

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:

$$C_6H_{12}O_6 + 6O_2 \Rightarrow 6CO_2 + 6H_2O$$

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:

$$C_6H_{12}O_6 \Rightarrow 3CO_2 + 3CH_4$$

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	1 810.03	

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:

$$G = Sr\rho 1/M_v X V_0$$

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_o molar volume of ideal gas (m^3/kmol)

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

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

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

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 $^{14}CH_4$ and $^{14}CO_2$. These gases result in radiation exposure of inhabitants living nearby repository. The effective dose of this critical group can be expressed as:

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

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

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

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

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

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 $(\text{Bq}\,\text{y}^{-1})$
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:

$$V_{\text{air}} = W u h t$$

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

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 $1.5 \times 10^{-6}\text{Sv}\,\text{y}^{-1}$. 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.

Acknowledgments

The technical and methodical support from Mr. Kövér and Mr. Remiáš (SE-VYZ) in data providing as well as encouraging discussion are gratefully acknowledged.

References

[1] BRUSH, L. H.: Test Plan for Laboratory and Modeling Studies of Repository and Radionuclide Chemistry for the Waste Isolation Pilot Plant, SAND90-0266, Sandia National Laboratories, New Mexico, 1990.
[2] BRUSH, L. H.: Systems Prioritization Method Iteration 2, Baseline Position Paper, Gas Generation in the Waste Isolation Pilot Plant, Sandia National Laboratories, 1995.
[3] KURUC, J.: Assessment of Gas Production and Pressure Conditions of FRC under Disposal Due to Microbial Degradation and Radiolysis of Cellulose, Mathematical model, OMEGA Info, Bratislava, 2002.
[4] Post-Closure Performance Assessment, Gas Generation and Migration, Science report No. S/94/003, United Kingdom Nirex Limited, November 1994.
[5] Derivation of Activity Limits for Disposal of Radioactive Waste to Near Surface Facilities, IAEA, Vienna, Austria 2001.
[6] WIBORGH, M.: Safety Assessment Methodologies for Near-Surface Disposal Facilities, Gas Generation and Transport. Kemakta Consultants Co.

Received 4 October 2005

Jozef Prítrský (Ing), born in Piešťany, Slovakia, in 1969, graduated from the University of Transport and Communication Žilina, Faculty of Mechanical and Electrical Engineering, Department of Mechanical Technology in 1991. At present he works in DECOM Slovakia, Ltd. as a senior expert in the field of radioactive waste and spent nuclear fuel management. His research interests include long term safety analyses of disposal facilities. Since 2003 he has been an external PhD student at the Department of Nuclear Physics and Technology, Slovak University of Technology, Bratislava.

Igor Matejovič (Ing, CSc), born in Piešťany, Slovakia, in 1958, graduated from the Faculty of Chemical Technology, Slovak University of Technology, Bratislava in 1982 and received the CSc degree in 1988 in plant sciences. At present he works in Decom Slovakia Ltd. Trnava as a project manager in the field of radioactive waste and spent nuclear fuel management and environmental impact assessment.

František Ondra (Ing), born in Liptovský Mikuláš, Slovakia, in 1979, graduated from the Slovak University of Technology Bratislava, Faculty of Electrical Engineering and Information Technology, Department of Nuclear Physics and Technology in 2003. At present he works in DECOM Slovakia, Ltd. as a junior expert in the field of standardized decommissioning costing and radiation safety analysis in decommissioning. He is an external PhD student at the Department of Nuclear Physics and Technology, Slovak University of Technology, Bratislava.

Vladimír Nečas (Prof, Ing, PhD), born in Banská Bystrica in 1954, graduated from the Faculty of Electrical Engineering, Slovak University of Technology, Bratislava in 1979 and received the PhD degree in 1990 in Experimental Physics. Since 1993 he has worked as Associate Professor and since 2001 as Professor at the Department of Nuclear Physics and Technology at the same university and faculty. At present the main fields of his research and teaching activities are experimental nuclear physics, nuclear reactor materials, problems of spent nuclear fuel and decommissioning of nuclear power plants.