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*Dedicated to Prof. Dumitru Barbu Ion's 70th Anniversary*

# SOME STORAGE ASPECTS OF SOLID RADIOACTIVE WASTES

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*Abstract*. The paper presents events and phenomena occurring during the storage of solid low level radioactive wastes that normally emerge from Cernavoda NPP. The GasSim model will be used to estimate the volume and pressure for the gases produced in a drum with compacted solid wastes by microbiological dispersion.

*Key words*: solid low level radioactive waste, radioactive waste storage.

## 1. INTRODUCTION

The low level solid radioactive wastes from CNE-Prod Cernavoda are segregated and collected in 220 l stainless steel standard drums, compacted and stored for about 15–20 years at the Radioactive Waste Intermediary Storage Facility. During the storage period, about 10% of all the drums appear to have swollen top covers. The deflection varies between 0 and 20 mm.

The causes for the swelling of the top cover of the drum are the biochemical transformations that occur in the bulk of the biodegradable waste, compacted and stored in closed spaces, due to the action of the anaerobic microorganisms. The phenomenon is known for some time and studied in detail for the last 15 years, because this is an essential issue in designing and constructing a radioactive waste disposal facility and also an ecological repository for domestic wastes.

In the case of storage or final disposal of low and intermediate level radioactive wastes the most important gas generation source is the degradation of cellulose based wastes like wood, paper, cardboard, textiles etc. due to the microbiological activity. Although in reality this degradation is done by different microbial populations, it is convenient and justifiable to describe it through a series of representative chemical reactions generated by 8 different microbial populations.

Microbiological activity degrades the cellulose based waste in the drum to soluble cellulose or polysaccharide ($[C_6H_{10}O_5]_n$), followed by the hydrolysis of this soluble form to a glucose based monomer ($C_6H_{12}O_6$). The resulting monomer acts like initial substrate for further degradation mediated by 8 different microbial populations. The microbial population depends upon the level of pH in the drum, the existence of nitrogen, water (as moisture) and some inhibitors [1].

The chemical reactions that model the waste degradation are:

$$\text{Bulk cellulose} \rightarrow \text{soluble cellulose (polysaccharide)} \tag{1}$$

$$[C_6H_{10}O_5]_n + nH_2O \rightarrow n\, C_6H_{12}O_6. \tag{2}$$

The further degradation of the glucose type monomer under the microbial attack:

1) Aerobic degradation (Microbe type 1)

$$C_6H_{12}O_6 + 6O_2 \rightarrow 6CO_2 + 6H_2O \tag{3}$$

The reaction occurs in the presence of oxygen fro the air trapped in the drum until it is consumed. After that only anaerobic reactions take place.

2) Nitrate reduction (Microbe type 2)

$$5C_6H_{12}O_6 + 24HNO_3 \rightarrow 30CO_2 + 12N_2 + 42H_2O \tag{4}$$

3) Anaerobic acidogenesis (Microbe type 3)

$$C_6H_{12}O_6 + H_2O \rightarrow 2CO_2 + 2H_2 + C_2H_5OH + CH_3COOH \tag{5}$$

4) Anaerobic acetogenesis (Microbe type 4)

$$C_2H_5OH + H_2O \rightarrow CH_3COOH + 2H_2 \tag{6}$$

5) Acetoclastic methanogenesis (Microbe type 5)

$$CH_3COOH \rightarrow CO_2 + CH_4 \tag{7}$$

6) Hydrogen-utilizing methanogenesis (Microbe type 6)

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \tag{8}$$

7) Acetoclastic sulphate reduction (Microbe type 7)

$$CH_3COOH + H_2SO_4 \rightarrow H_2S + 2CO_2 + 2H_2O \tag{9}$$

8) Hydrogen-utilizing sulphate reduction (Microbe type 8)

$$4H_2 + H_2SO_4 \rightarrow H_2S + 4H_2O \tag{10}$$

Physical degradation of the cellulose from the solid wastes to soluble cellulose or polysaccharide followed by the hydrolysis of this soluble form produces a monomer. The hydrolysis takes place both naturally and microbial mediated. Henceforward the 8 microbe types act in stages including both aerobic and anaerobic regime and the two regimes are not clearly separated in time and space. While air is available the dominant reaction is reaction (3), the aerobic degradation of the waste under the action of the type 1 microbe. When oxygen is depleted the anaerobic populations become increasingly active and, as nitrate reducers (type 2), sulphate reducers (type 7 and 8), acidogens (type 3), acetogens (type 4) and methanogens (type 5 and 6). The microbial populations are influenced by the level of pH.

## 2. GAS VOLUME ASSESSMENT

The assessment of gas volume produces in drums with cellulose based waste, stored at Radioactive Waste Intermediary Storage Facility will be done based upon literature data [2].

For a better assessment of the generated gas volume the cellulose based waste are classified in 3 categories, the degradation rate under microbial attack being the classification criteria, and presented in Table 1.

*Table 1*

Degradation rate for types of cellulose based wastes [2]

| Degradability | Waste type | Degradation time |
|---|---|---|
| Rapid | Putrescibiles<br>Garden waste<br>Sewage sludge<br>Fines | 6 years |
| Moderate | Paper (aprox. 25%) – except newspaper<br>Nappies<br>Miscellaneous combustibles | 9 years |
| Slow | Paper (aprox. 75%) – except newspaper<br>Newspaper<br>Textiles | 15 years |

GasSim model is used for the assessment of the generated gas volume, this model being used by the United Kingdoms Environment Agency.

Validation of the model was validated was done with gas volume measurements. The gas volume was generated through the degradation of known amount of waste and it is an exponential function and it is time dependent [2].

The calculated gas generation capacity was assessed at 64 $m^3$/tone of waste, in the climateric conditions from United Kingdom.

The GasSim model leads to a time dependent gas generation rate which is close to the experimental curve and it is presented in Fig. 1.

The gas volumes produced through microbial degradation cumulated for a 150 years period are presented in Table 2.

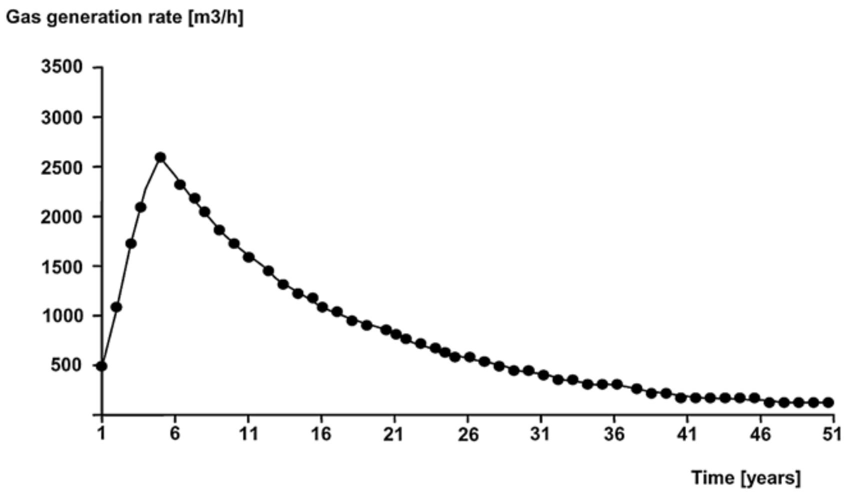


Fig. 1 – Time variation of the gas generation rate [2].

*Table 2*

Cumulated gas volume produced through microbial degradation in 150 years [2]

| Model | $CH_4$ ($m^3$/t) | $CO_2$ ($m^3$/t) | Total gas volume ($m^3$/t) |
|---|---|---|---|
| GasSim | 63.7 | 63.7 | 127.5 |

## 3. RESULTS AND DISCUSSIONS

The assessment of the gas generated in drums containing compacted waste at Cernavoda NPP the data obtained with GasSim model will be used. Thus the curve from Fig. 1 will be normed to a tone of cellulose based wastes and will result the curve presented in Fig. 2.

In the period between 1 and 6 years the generation rate can be assessed using time linear function [3]:

$$V(t) = \text{const} \cdot t \tag{11}$$

Where the constant has the value of the tangent of the slope angle of the curve relative to the time axis and it can express as:

$$\text{const} = \frac{6.0 \cdot 10^{-4}}{5 \cdot 24 \cdot 365} \left[ \frac{\text{m}^3}{\text{ton} \cdot \text{h}} \right] \tag{12}$$

In order to know total volume of accumulated gas during the 5 years of waste degradation, the function $v(t)$ is integrated for a 5 year time period [3].

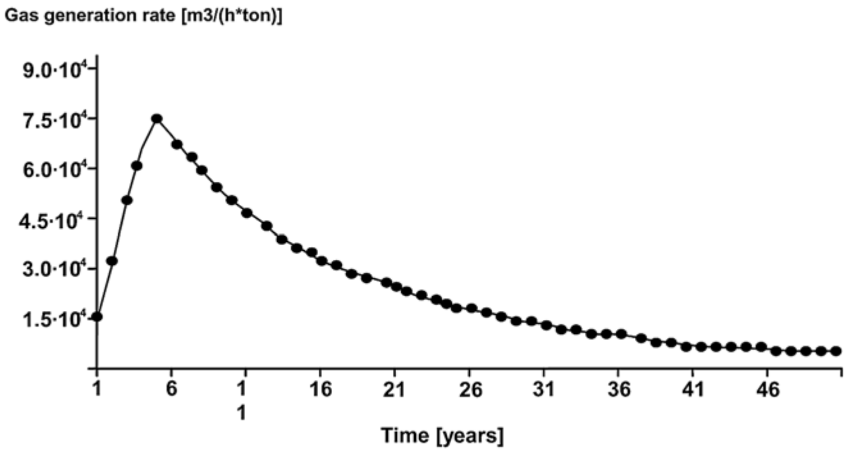


Fig. 2 – The variation curve of the assessed, with GasSim, gas generation rate [3].

$$V(t) = \frac{6 \cdot 10^{-4}}{5 \cdot 24 \cdot 365} \int_{0}^{5 \cdot 24 \cdot 365} t dt \tag{13}$$

The calculated gas volume is 13.14 $m^3$/tone.

The average net mass of the drums with compacted cellulose based waste is 95 kg, so the gas volume that will be produced by the waste in a drum over a period of 5 years is 1,248 $m^3$.

Considering the gas volume determined using GasSim model for a tone of cellulose based waste for a period of 150 years (Table 2), the gas volume produced in the bulk of the waste (95 kg) in a drum it can be calculated:

$$\frac{127.5 \cdot 95}{1100} = 12.12 \text{ m}^3 \tag{14}$$

From Fig. 2 it can be observed that, practically, after 50 years, which represent ten time periods of maximum gas generation rate, the whole quantity of cellulose based waste is degraded under the microbial attack and in an acceptable approximation, the produced gas volume is 1.211 $m^3$.

The two values of generated gas volume from the cellulose waste from a drum over a 5 years period, after compacting are in good concordance.

## 4. GAS PRESSURE ASSESSMENT

After 5 years of storage, in a drum with cellulose based wastes, a gas volume of 1,248 $m^3$ is generated through microbial degradation. The gas will produce an overpressure in the drum, resulting in swelling of the top cover.

In order to evaluate this overpressure we will consider the drum without wastes at atmospheric pressure $p_0$ and with volume $V_0$ = 220 l.

The perfect gas equation in this case is:

$$p_0 \cdot V_0 = n_0 \cdot R \cdot T \tag{15}$$

where: $p_0$ = pressure;
$n_0$ = number of air moles in the drum;
$R$ = perfect gas constant;
$T$ = temperature [K];
$V_0$ = drum volume, $V_0$ = 220 l.

Considering that in the drum 1.248 m$^3$ of gas is introduced the perfect gas equation in this case becomes:

$$p_0 \cdot V_1 = n_1 \cdot R \cdot T \tag{16}$$

where: $p_0$ = 1 tam;
$n_1$ = number of moles;
$V_1$ = 1248 l.

After the gas is introduced in the drum, the number of moles in the drum will be $n_0$ + $n_1$, and the pressure will be $p_x$. So the perfect gas equation becomes:

$$p_X \cdot V_0 = (n_0 + n_1) \cdot R \cdot T \tag{17}$$

$$n_0 = \frac{p_0 \cdot V_0}{R \cdot T} \tag{18}$$

$$n_1 = \frac{p_0 \cdot V_1}{R \cdot T} \tag{19}$$

Taking into account equations (18) and (19) in equation (17) will result:

$$p_X = p_0 \left( \frac{V_0 + V_1}{V_0} \right) \tag{20}$$

$$p_X = 6{,}67 \cdot p_0 \tag{21}$$

In the real case, when the drum is filled mostly with waste, the free volume after the waste is compacted will bee filled with gas. This free volume is maximum 15% of the total volume of the drum. Considering $V_0$ = 33 l in equation (20) will result:

$$p_X = 393 \ \frac{\text{N}}{\text{cm}^2} \tag{22}$$

Thus through the microbial degradation of the compacted cellulose based waste in 220 l stainless steel drums a maximum pressure of 400 N/cm$^2$, uniformly distributed on the cover, can be achieved.

The drum behavior at the overpressure due to the gas generated through microbial action is presented bellow.

The deflection of the cover of the drums containing compacted cellulose based waste varies between 0 and 10 mm. This deflection is due to the gas produced during the degradation of the waste under microbial attack.

In order to determine the necessary force to cause a cover deflection of 10 mm, we use the formula for the calculus of the deflection value of a circular plate circular embedded, with a uniformly distributed load [5]:

$$f = \frac{3 \cdot P \cdot \left(m^2 - 1\right) \cdot a^2}{16 \cdot \pi \cdot m \cdot E \cdot h^3} \tag{23}$$

where: $f$ = deflection ($f = 10^{-2}$ m);

$h$ = plate width ($h = 1.5 \cdot 10^{-3}$ m);

$P$ = uniformly distributed load (pressure);

$m$ = Poisson coefficient ($m = 3$);

$a$ = plate radius ($a = 0.3$ m);

$E$ = coefficient of elasticity (stainless steel 304L, $E = 2.1 \cdot 10^{11}$ N/m$^2$)

From equation (23) results:

$$P = \frac{16 \cdot \pi \cdot m \cdot E \cdot h^3 \cdot f}{3 \cdot \left(m^2 - 1\right) \cdot a^2} \tag{24}$$

From this data we obtain $P = 494.5$ N, which represents the pressure on the cover of the drum.

The stainless steel drum underwent the free fall test in order to evaluate the shock behavior and also to evaluate the strength of the locking ring of the cover. For the free fall test the sample drum is filled with sand and it is let to fall freely on one of the tops from a height of 1.2 meters [4].

From the tests done, both type test and batch test, there were observed deformations, of both the cover and the locking ring, of maximum 6 mm. In order to evaluate the cover strength we will use the kinetic energy variation theorem.

The variation of the kinetic energy in the time interval $dt$ is equal to the mechanical work done in the same time interval: $\delta E = L$.

$$\delta\left(\frac{mv^2}{2}\right) = \vec{F} \cdot \delta\vec{r} \tag{25}$$

where: $E$ = kinetic energy, $E = \frac{mv^2}{2}$;

$L$ = mechanical work done for cover deformation;

$m$ = drum mass;

$v$ = free fall rate of the drum;

$\vec{F}$ = uniformly distributed force on the cover;

$\delta\vec{r}$ = variation of the positioning vector $\vec{r}$.

In order to calculate the free fall rate $v$, at the impact moment, we can use the formula for free fall in gravitational field:

$$H = \frac{g \cdot t^2}{2} \tag{26}$$

$$v = g \cdot t \tag{27}$$

where: $H$ = height;

$g$ = gravitational acceleration;

$t$ = free falling time from 1.2 m.

From equation (26) results:

$$t = \sqrt{\frac{2H}{g}}\ . \tag{28}$$

From this data we obtain the value of the free falling rate, at the impact moment, $v = 4.85$ m/s.

The sum of the mass of the empty drum and the mass of the sand is 345 kg. So the kinetic energy variation can be calculated:

$$\delta\left(\frac{mv^2}{2}\right) = 4163\ \ \text{kg}\,\frac{\text{m}^2}{\text{s}^2} \tag{29}$$

for $\delta(\vec{r}) = 0{,}006$ m, from equation (25), we obtain the force $F = 6{,}93 \cdot 10^5$ N.

From data comparison we can estimate that the drum will resist without problems to the stresses due to the gas generation from waste degradation inside the drum, over the whole period of waste storage.

## 5. CONCLUSIONS

The deflection of the stainless steel drums cover used for storing solid radioactive waste, which contain cellulose based materials is due to the gas produced during the organic materials degradation under microbial attack.

The microbial degradation scheme of the organic materials is reduced to three general forms of chemical reactions.

The maximum amount of gas generated is at maximum 6–8 years after the drum with cellulose based waste is closed.

The maximum pressure which stresses the cover and it is responsible for the cover deflection is 393 N/cm$^2$.

## REFERENCES

1. Science Report, *Post Closure Performance Assessment. Gas Generation and Migration,* United Kingdom NIREX Limited, Report No. S/94/003, 1994.
2. R. Attenborough, D. Gregory, D. Hall, L. Mac Goochan, *Development of a Landfill Gas Risk Assessment Model: Gas Sim*. Environmental Agency, West Malling England.
3 I. V. Popescu, *Analiza cauzelor și a consecințelor deformării capacelor butoaielor de 220 l ce conțin deșeuri solide presate de la CNE Cernavodă*, Raport intern, Pitești, 2006.
4. E. Dumitru, *Stocarea deșeurilor radioactive. Realizări și perspective, principii și legislație,* Referat 3, Pitești, 2005.
5. P. Augustin, C. Constantin, *Manualul inginerului*, Vol. 2, Editura Tehnică, București, 1995.