Conf-910485--49

POTENTIAL $^{14}CO_2$ RELEASES FROM SPENT FUEL CONTAINERS AT YUCCA MOUNTAIN

BNL-45658

Received by OSTI

BNL--45658

DE91 008091

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ABSTRACT

The potential release of gaseous $^{14}CO_2$ from small perforations in spent fuel containers has been evaluated as a function of temperature, hole size, effective porosity of corrosion products within the hole, and time, based on the waste package design parameters and environmental conditions described in the Yucca Mountain Site Characterization Report (SCP). The SCP does not specify initial fill gas (argon) pressure and temperature. It is shown that, if significant ^{14}C oxidation takes place during the initial, inert-gas phase, an incentive exists to initially underpressurize the containers. This will avoid large, spiked releases of gaseous $^{14}CO_2$ and will result in delayed, smaller, and more uniform release rates over time. Therefore larger size perforations could be tolerated while meeting the applicable regulations.

INTRODUCTION

In recent years, the potential release of ^{14}C from failed spent fuel containers at Yucca Mountain has become an important licensing concern [see reference 1 for a detailed list of references]. This is due to the very small releases which the Environmental Protection Agency (EPA) and the Nuclear Regulatory Commission (NRC) high-level waste regulations allow.

According to the EPA rule presently under review, the *cumulative* release of ^{14}C from the repository to the accessible environment should not exceed 0.1 Ci per metric ton of heavy metal (MTHM) over 10,000 years after disposal[2]. This corresponds to a maximum cumulative release of 7,000 Ci, i.e., an average rate of release of 0.7 Ci/yr[a]. The latter is a very small rate considering that ^{14}C is likely to be released from spent fuel in gaseous form[3], mostly as $^{14}CO_2$, and that, according to German data[4], routine releases of gaseous ^{14}C compounds to the environment from the stacks of nuclear power plants amount to roughly 5.6 Ci/yr for 1 GW(e) PWRs and 13.5 Ci/yr for 1 GW(e) BWRs.

The NRC rule[6] limits the release *rate* of ^{14}C during the controlled release (CR) period to no more than 1 part in 10^5 per year of the ^{14}C inventory at 1,000 years or, alternatively, to no more than 1 part in 10^8 of the total curie inventory of the waste present at 1,000 years. According to the first criterion, considering a ^{14}C inventory at 1,000 years of about 1.37 Ci/MTHM (based on PWR fuel with 33,000 MWd/MTHM burnup and BWR fuel with 27,500 MWd/MTHM burnup[7])[b] and a 70,000 MTHM[a] of spent fuel waste to be disposed of, one obtains a maximum release rate of roughly 1 Ci/yr. Based on a total waste inventory of 1,730 Ci/MTHM at 1,000 years, the alternate criterion allows instead a maximum release rate from the repository of 1.2 Ci/yr.

A third and more stringent criterion is the NRC's requirement of substantially complete containment (SCC) of all radionuclides within the waste package for a minimum period of time between 300 and 1,000 years after repository closure. No numerical statement of allowable releases during the SCC period is provided. It has been proposed by the

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[a] The waste acceptance criteria in the DOE Mission Plan[5] specify only 63,000 MTHM of spent fuel with the remaining 7,000 MTHM in glass waste forms. ^{14}C release from glass waste forms is believed to be negligible, therefore, use of 70,000 MTHM is conservative as it overestimates inventories by 10%.

[b] More recent estimates by Van Konynenburg[8], indicate a ^{14}C activity of 0.97 Ci/MTHM at 1,000 years.

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DOE that a further tightening of the controlled release rate limits by one order of magnitude may be acceptable. This would result in a maximum, allowable release rate of roughly 0.06-0.1 Ci/yr from the whole repository during the SCC period.

Assuming, for the sake of simplicity, that all waste packages contain 6 PWR 17x17 spent fuel assemblies, one obtains a ^{14}C inventory of 4.285 Ci (based on Roddy's inventory data[7]) per container at time of disposal and a total number of 25,285 containers. Thus the allowable yearly releases from the whole repository correspond to roughly one fortieth or *less* of the initial ^{14}C inventory of a *single* container and drive container design considerations.

In calculating the actual release rates from a repository at Yucca Mountain one may take into consideration that:

1. all of the ^{14}C inventory may not be available for release at time of breach or later;

2. there may be significant mass transfer resistance to release through the openings in the container wall; and

3. there will be a time distributed failure rate of spent fuel containers.

The present communication addresses some of the issues relevant to item 2. Our main conclusion is that, if significant oxidation of the ^{14}C takes place during the initial, inert-gas phase, underpressurization of the original spent fuel containers should be considered in order to help meet the regulatory criteria and to ensure relatively slow, uniform releases in case of container breach.

REFERENCE DESIGN DATA AND ENVRONMENTAL CONDITIONS

In order to examine the mass transfer problem of ^{14}C through openings in waste containers, a typical spent fuel container loaded with intact fuel waste is envisioned with relevant properties taken from the Yucca Mountain Site Characterization Plan[9] (SCP).

The reference container has a wall thickness, ℓ, of 1 cm with a free volume inside, V, of 1 m^3. The initial fill gas is argon. In our calculations the viscosity of argon as a function of temperature was represented as follows[1]:

$$\mu(T)=208.72+0.9378\cdot T-0.1299\cdot T^{1.2}$$

with $[\mu]=[\text{micropoise}]$ and $[T]=[^{o}C]$. The self-diffusion coefficient of argon, estimated from Fuller's correlation[10] was expressed as follows:

$$D(\vartheta,P) = 8.836\cdot 10^{-6}\cdot \frac{\vartheta^{1.75}}{P},$$

with $[\vartheta]=[^{o}K]$ and $[P]=[atm]$ and $[D]=[cm^2/s]$.

Based on the Yucca Mountain SCP, the anticipated temperature history of the containers' walls was interpolated as follows:

$$T_w(t)=92.774-0.0189\cdot t+359.08\cdot t^{-0.3}$$

with $[T_w]=[^{o}C]$ and $[t]=[yr]$. This temperature curve only applies to the hottest containers for $t \geq 15$ years. Containers placed farther away from the center of the repository are expected to exhibit a lower initial temperature increase (edge effects). The validity of the above correlation cannot be extended much beyond its range of interpolation of the SCP data, 1,500 years say, as it may yield unrealistic results. However, it is the relatively early failures that cause most concern both because the release rate criteria are strictest at those times and because the oxidation rate of ^{14}C in air is larger the higher the temperature[11].

No internal temperature calculations of the *present* SCP waste package designs are available[12]. Thus the *average* temperature of the gas within the waste container, which controls the fill gas pressure, is not known. By examining the more detailed work performed in the allied field of dry storage of spent fuel[12], we have *estimated* that the ratio of the average gas temperature expressed in degrees Kelvin, ϑ_b, to the wall temperature expressed in degrees Kelvin, ϑ_w, should be of the order of 1.1 or less. Accordingly, in our calculations we approximated $\vartheta_b(t)$ as:

$$\vartheta_b(t) = 1.1\ \vartheta_w(t)\ .$$

The SCP does not indicate a reference argon fill pressure or temperature. These were left open as design variables. In any event, we have assumed that enough argon is utilized that the oxidized ^{14}C fraction available for release at all times can be considered as an impurity in the fill gas. This assumption is justified by noticing that

even if the total ^{14}C inventory of a container (4.285 Ci) were to be oxidized into $^{14}CO_2$ its partial pressure would amount to only 0.002 atm at room temperature and that, in practice, only a few percent of the initial ^{14}C inventory will be present as $^{14}CO_2$ in a container at any time.

MODELING APPROACH

In order to be able to model the ^{14}C release from failed spent fuel containers we subdivided the problem into two time domains associated with different flow regimes. In the first time domain the internal fill gas (argon) pressure, assumed to be higher than the outside pressure, tends to equilibrate with the external pressure. The flow regime is essentially of the Poiseuille type for compressible gases during which the available $^{14}CO_2$ is carried outwards as an impurity in argon. As soon as equilibration is achieved, air is assumed to rapidly flow into the container and serve as the new fill gas. At this time the new time domain begins. The assumption that air becomes the new fill gas upon pressure equilibration is conservative because it provides an instant source of oxygen to oxidize the ^{14}C on the cladding. In this second phase we are interested in the interplay between the outward diffusion of a gaseous impurity, i.e., $^{14}CO_2$, and the inflow of air needed to compensate for the internal pressure decrease due to the decaying temperature field (counterdiffusion regime).

In the first time domain the inwards diffusion of outside oxygen (which could further oxidize the ^{14}C inventory of the cladding) against the argon outflow from the container was neglected, because our calculations suggest that, during this early phase, the argon outflow velocity is high enough to practically annul this counterdiffusion process.

The original mathematical equations that were developed for our calculations are provided in a companion report of ours[1].

In our calculations for both time domains we factored in the effective porosity, ϵ, of the perforation and the associated tortuosity, τ, of the flow path. Thus we could model the release from totally open, $\epsilon=1$, and from partially occluded, $\epsilon<1$, perforations. Indeed, localized corrosion is regarded as the most likely failure mode of spent fuel containers in a wet or humid environment and since, for iron corrosion products, the molar volume exceeds that of the base metal (Table 1), the potential exists for the corrosion products to form a well-packed barrier to flow. Furthermore, small penetrations could also become plugged with particulates deposited by the outflowing and inflowing gases.

Table 1 Molar volume [cm³] of iron and its common corrosion products (Adapted from CRC Handbook of Chemistry, 1981).

Compound	Specific Volume per mole of Fe
Fe	7.11
Fe_3O_4	14.9
$Fe(OH)_2$	26.4
FeO(OH)	20.8
Fe_2O_3	15.25

The available, oxidized fraction of the total ^{14}C inventory at time of breach, f, is temperature dependent and was taken to be as follows[1]:

$$f = \begin{cases} 2\% & \text{for } T > 215\ ^{o}C \\ 1.2\% & \text{for } T < 215\ ^{o}C \end{cases}$$

These are more conservative values than those suggested in the Yucca Mountain SCP.

The treatment of porosity and tortuosity effects during both the pre- and post-equilibration time domains represents an improvement on earlier modeling work[15,16] in the literature. A further improvement is represented by the incorporation in our models of the air inflow velocity as function of temperature during the post-equilibation phase. In particular, the mass flow velocity, u, dependence on temperature (in degree K), gas volume, perforation radius and porosity is as follows[1]:

$$u = \frac{V}{\pi \epsilon r^2} \cdot \frac{1}{T} \cdot \frac{\partial T}{\partial t}$$

RESULTS

Figure 1 represents a typical curve for the fractional release rate versus time when the release is controlled by the pressure-

driven flow of the argon gas out of the container (phase 1). With a fill gas of 1 atm at 25 °C, it shows a marked, spiked release which barely meets the controlled release criteria[c] of the NRC (if all containers had the same type of breach) even if the perforation radius is a mere 5 μm and the available, oxidized carbon fraction is only 1%. Larger size perforations will result in larger releases while, at the same time, the equilibration times become shorter. In particular, as shown in Table 2, equilibration to 1 atm will take place in less than a year for 35 μm radius perforations and larger. By the time equilibration of the inside and outside pressure is achieved 30 to 50% of the argon gas and of the $^{14}CO_2$ inventory are released.

Table 2 Time-to-pressure-equilibration and fractional amount of gas released as function of different hole radii and breach times.

radius [μm]	t_{start} [yr]	Δt_{eq} [yr]	$\Delta n/n_o$ [%]
	15	855	31.7
5	300	900	29.8
	600	900	28.2
	15	76	40.7
10	300	100	35.1
	600	104	32.7
	15	1.0	48.1
35	300	1.0	36.2
	600	1.0	33.4

Figure 2 represents the fractional release rates from a 50 μm radius open perforation in which outwards diffusive release of $^{14}CO_2$ occurs against the advective inflow of air (phase 2). For this size perforation, equilibration of the inside and outside pressure is practically instantaneous, but an advective flow still exits in order to offset the decay of the internal pressure due to the decrease in temperature within the container. In this figure we do not address the mass loss during pressure equilibration, i.e., we look at a situation where the container is at the same pressure as outside from the start of breach. The oxidized ^{14}C fraction is 2% for the 15- and 100- years breach times and 1.2% otherwise. It is to be noted that in all cases, the fractional release rate is now less than $1.2 \cdot 10^{-5}$, which is lower than the NRC release rate limit of $1.8 \cdot 10^{-5}$.

Comparing Figure 1 and Figure 2 with each other, one readily realizes they are strikingly different, the latter showing no spiked release even for a substantially larger perforation and oxidized carbon fraction. The release curves for breach at 15 and 100 years are especially interesting as they show lowest release rates at early times. This is because the rate of decrease in the internal temperature of the spent fuel container is largest initially causing a significant inflow of air. This effect is important for perforation radii of up to 150 μm. For larger radii, a slowly decreasing release rate behavior similar to the lower curves of Figure 2 will prevail.

Underpressurization

Because of the above findings we believe it would be worthwhile to cause releases to be controlled by diffusion against a counterflow of air at all times. This could be accomplished by filling the container to a pressure such that, at the peak average fill gas temperature, it would not exceed the outside pressure. As the system cools, the internal pressure would decrease further, ensuring that upon breach, flow would be into the container.

Even though it may seem counterintuitive to permit oxygen to flow into the container initially, it will not have a detrimental effect on release for the following reasons. First, for a perforation with a radius of 35 μm or larger, pressure equilibration occurs in 1 year or less. Thus, even if the container is at a higher pressure than the surrounding environment, delay of oxygen entering the container is at most a year. This depressurization step would be characterized by a spiked release, while underpressurization would promote a slow, uniform release. Second, for a smaller radius perforation, e.g., 5 μm radius, if the container is underpressurized with respect to the outside

[c] Assuming a total waste inventory at 1,000 years of $1.74 \cdot 10^3$ Ci/MTHM[7] and the most recent estimate of 0.97 Ci/MTHM for the ^{14}C inventory at 1,000 years[8], it is seen that a fractional release rate per container of roughly $1.8 \cdot 10^{-5}$ would be able to meet the total inventory release rate limit of the NRC.

environment, ^{14}C release will occur against a very significant counterflow of air which will cause the NRC release criteria never to be exceeded.

Filling to a lower pressure would utilize less argon, an expensive noble gas, but it may have detrimental effects on other parameters relevant to waste package performance, e.g., heat transfer, leak detection, and ease of fabrication. Decreasing the amount of fill gas in the container will certainly alter convective heat transfer; however, to date, none of the thermal analyses of spent fuel waste packages have considered convection[12]. Filling to a lower pressure will not allow sniffing for a gas leak; however, it should not influence ultrasonic and liquid penetrant tests, which "... *represent the best choices for closure certification*" [14].

The ease of fabrication for a spent fuel waste container may be adversely affected if underpressurization is used. For example, friction welding may no longer be considered and it may prove difficult to maintain a partial vacuum within the container and in the hot cell. Other possible consequences of having a lower fill gas pressure should be examined before proceeding.

Although we recommend underpressurization as a means of reducing the pulse type of release associated with pressure equilibration, the advantage gained in underpressurizing the container may be significantly reduced if the oxidized fraction of ^{14}C is significantly less than our assumed values in an argon environment. Release is proportional to the oxidized fraction.

Meeting Potential SCC Release Limits

Limiting gaseous releases by diffusion against air flowing into the container also offers some hope in demonstrating that substantially complete containment can be met. DOE has proposed to reduce release rate limits by a factor of 10 during the substantially complete containment period. This allows two possible release rate limits: a) 10^{-6} of the ^{14}C 1,000 year inventory, and b) 10^{-9} of the total 1,000 year inventory which is $1.8 \cdot 10^{-6}$ of the ^{14}C 1,000 year inventory.

We have estimated the *cumulative* fraction of failed containers that could be tolerated in order to meet those release limits. Our results are presented in Table 3 which provides the cumulative percentage of failed containers as a function of release rate limit, hole size and porosity.

TABLE 3 Maximum cumulative fraction of failed containers as a function of hole size, porosity (ϵ), and fractional release rate (FRR) limit.

Radius [μm]	Max % failed FRR=10^{-6}	Max % failed FRR=$1.8 \cdot 10^{-6}$
	$\epsilon = 1$	
50	8 - 13	14 - 23
80	3 - 5	5.4 - 9
100	1.5 - 3	2.7 - 5.4
	$\epsilon = 0.1$	
100	50 - 100	100
500	3.5 - 5.5	6.3 - 10
1000	0.8 - 1.4	1.4 - 2.6
	$\epsilon = 0.01$	
500	100	100
1000	33 - 50	60 - 90

The range of values for the maximum fraction of failed containers reported in Table 3 is connected to early or later failure times. For failures at less than 100 years, the fraction of oxidized ^{14}C is assumed to be 2%, after 100 years it is 1.2%. Lower failure rates are necessary for breach times less than 100 years.

From Table 3 it can be established, for instance, that if up to 3% of the waste containers were to fail during the SCC period, the DOE-suggested release criteria would be met provided the perforations radii were not larger than 100 μm, say, for fully open perforations, and not larger than about 1 mm, say, for partly occluded perforations with 10% effective porosity.

CONCLUSIONS

The potential release of gaseous $^{14}CO_2$ from small perforations in waste containers has been evaluated as a function of temperature, hole size, open porosity within the hole, and time, based on the waste package design parameters and environmental conditions described in the Yucca Mountain Site Characterization Report.

The analysis has been performed by breaking the release problem into two time domains. In the first time domain a positive ΔP between the argon fill gas and outside air drives the former out of the spent fuel container (type-1 conditions). The flow regime is essentially of the Poiseuille type for compressible gases. By the time equilibration of the inside and outside pressure is achieved, 30 to 50% of the argon gas and of the $^{14}CO_2$ inventory (modeled as an impurity in the argon gas) are released. Equilibration will take less than a year for 35 μm radius perforations and larger. For these perforations the release of $^{14}CO_2$ can be characterized as a spiked release. Following pressure equilibration, air is assumed to be the new fill gas instantly oxidizing the exposed ^{14}C. In this time domain $^{14}CO_2$ counterdiffuses against the inflow of air leaking into the container in order to offset the natural decrease in pressure due to the decay in the temperature field (type-2 conditions). The inflow of air is significant for perforations of 100 μm radius and smaller, and it is most important at early times. This type of flow regime is characterized by relatively uniform releases over time. In both time domains we have utilized conservative assumptions regarding modeling itself and the environmental parameter values to be used, e.g., we considered a temperature-versus-time profile more typical of the hottest containers, whereas it is known that edge effects at the time of emplacement will cause smaller temperature increases.

The $^{14}CO_2$ release models that were developed can and should be used for analyzing the potential of present container designs to meet the EPA and NRC regulations. An important conclusion already apparent from the present work is that, if indeed significant oxidation of the ^{14}C takes place during the inert gas phase, an incentive exists to originally underpressurize the containers so that a negative ΔP will always exist at time of breach. This will ensure that type-2 conditions will prevail avoiding spiked releases, *removing* small-size perforations (50-μm radii and smaller, say) from the list of potential offenders, and resulting in the smallest releases at early times when the release criteria are strictest.

Alternatively, our analysis helps provide the basis for requesting changes in the regulations if it is found impossible to demonstrate with reasonable assurance that the perforation area will be limited to such low levels as the present regulations require. This might prove eventually to be the case given the current state-of-the-art in modeling corrosion processes and given the foreseeable uncertainties in establishing the potential environment to which the spent fuel containers will be exposed.

In any event, it is recognized that the release rate limits on ^{14}C from a HLW repository are extremely stringent and not in line with the criteria applicable to other phases of the nuclear fuel cycle. Therefore, it would be reasonable to request a relaxation of the EPA release criterion for ^{14}C by one order of magnitude or slightly more. This would allow release rates from a repository to be similar to those from nuclear power plants.

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This work was performed under the auspices of the United States Department of Energy.

Figure 1 Fractional ^{14}C release rate from a 5-μm radius, fully open perforation with breach occurring at 15 years after closure of the repository. The fill gas pressure is 1 atm at 25 °C. The available ^{14}C fraction in the form of $^{14}CO_2$ is 1% of the total ^{14}C inventory.

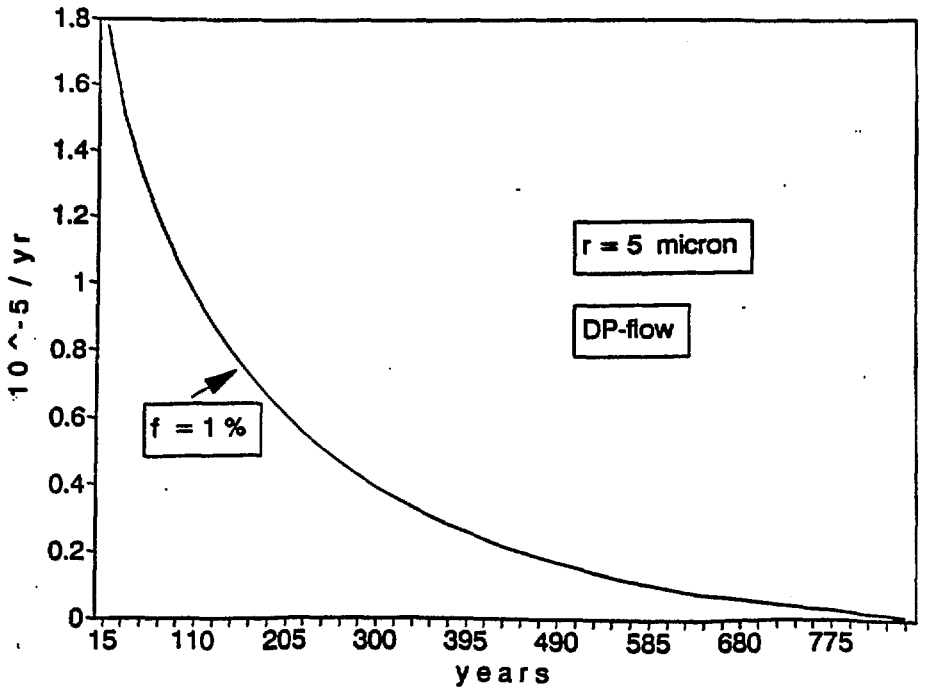
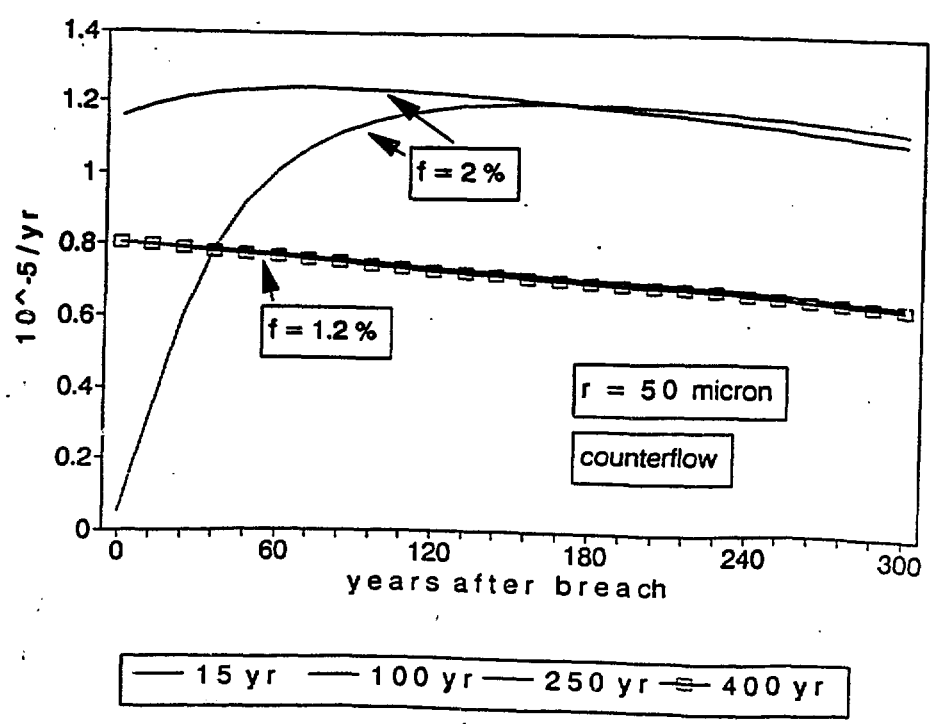
Figure 2 Fractional ^{14}C release rates from a fully open perforation of 50-μm radius assuming pressure equilibration conditions and with breach occurring at 15, 100, 250, and 400 years. The available ^{14}C fraction in the form of $^{14}CO_2$ is 2% of the total ^{14}C inventory for breaches formed before the year 120 from repository closure and 1.2% thereafter.