STORAGE OF TRITIATED WATER IN SALT CAVERNS

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ABSTRACT

Salt caverns kept at a relatively low internal fluid pressure by a permanent hydrocarbon blanket offer an appealing solution to the problem of hazardous-waste disposal — especially for radioactive liquid waste with a relatively short half-life (e.g., a decade). In such a system, there is little groundwater flow, and it is directed toward the cavern, preventing any waste seepage. The stored waste is contained safely (even in the case of well-head failure) and can be retrieved at any time. Cavern convergence is slightly accelerated by reducing the density of the fluid column in the well provided that cavern depth is not too large. Finally, much experience has been gained from the operation of thousands of existing salt caverns.

INTRODUCTION

Tritium (T) is a by-product of many nuclear reactions. It is a radioactive isotope of hydrogen, with a mass about three times that of ordinary hydrogen. In general, it is combined with water, and the so-called tritiated waters are a mixture of H_2O, THO and TO_2 molecules.

The basic disintegration of Tritium results in Helium and the emission of β-radiation (i.e., electrons). The corresponding amount of radiation is not very sharp —it can be stopped by a sheet of paper. However, direct contact with the skin, or ingestion, can lead to severe health problems. The activity of 1 gram of Tritium is 359 TBq (or 9700 Ci), but this figure does not give a clear image of the toxicity of Tritium, as it must be multiplied by a coefficient that takes into account its small radiated energy per gram and its short biologically active period. For Tritium, this coefficient is exceptionally small. From a practical point of view, a radioactive dose of 1 mSievert per year is generally considered to be a reasonable limit. For example, this dose would be reached by a person who drinks tritiated water with a concentration of 2×10^{-7} Ci/liter for an entire year.

Tritium can be extracted easily from waters with concentrations higher than 2000 Ci/liter. At the present time, tritiated waters with concentrations smaller than the above figure are released into the atmosphere or released in the ocean at locations where the waters are stirred by natural streams.

Nevertheless, due to the increasing concerns for environmental protection, there is mounting pressure to reduce the amount of radionuclides released in the biosphere to zero. Whether such action is sound is not the topic of this paper. Reducing the amount of radionuclides released into the atmosphere to zero leads to concentration of the wastes in a small volume of high toxicity, rather than in the dilution of waste in such a large volume of water or air that the resultant activity per unit volume of fluid becomes negligible. It implies that confinement of the concentrated wastes be extremely effective; seepage to the biosphere must be nil, or so small or so slow, that the released amount of radionuclides will not be harmful.

TRITIUM PERIOD

The half-life of Tritium is 12.43 years, which means that its activity (then, its toxicity) naturally decreases by 5% per year. In other words, the initial activity of Tritium is divided by 250 after 100 years and by 62,500 after 200 years. One very important characteristic of tritiated medium-activity waste is that it must be removed from the biosphere during a period of time which, albeit long, is limited. In sharp contrast to high-level nuclear waste storage, the disposal of tritiated waters relies on known storage techniques that can be assessed via full-scale experiments.

The major objectives for tritiated-water storage can be summed up as follows.

1. The storage must remain safe for a period of, typically, 100 to 200 years.

2. The required volume is several thousands of cubic meters.

3. The handling of waste during storage operations must be as simple as possible. Risk of pump failure, water leaks and more generally hazardous events must be minimized for all personnel involved.

4. The tritiated waters released from the storage must be nil.

The last statement is somewhat too stringent; the release of small amounts of radionuclides would not be harmful provided that seepage rates are small and that the released quantities were diluted in a very large volume of water before reaching the biosphere. However, it is important to prove that fluid seepage is nil, both through theoretical considerations and in-situ testing. Such a demonstration would help build public confidence.

BASIC STORAGE PRINCIPLE

The objectives cited above can easily be met by storing tritiated waters in salt caverns, which differ from standard oil- or gas-storage caverns in that the fluid pressure in the cavern is kept lower than the "halmostatic" pressure — i.e., the pressure of a cavern whose well is filled with saturated brine and opened to the atmosphere at ground level.

Much experience with leaching and operating storage salt caverns is available for study. The depths of such caverns range between 500 m and 2000 m. They are leached out from a salt formation. Typically, a 1-km deep vertical well is cased and cemented to the rock formation, with its shoe anchored to the top of the salt formation. A smaller central tube allows soft water to be injected at the bottom of the cavern. After the soluble rock salt is leached out, brine is removed from the cavern through the annular space between the cemented casing and the central injection tube, and a 10,000-m^3 to 1,000,000-m^3 cavern is created after about a year's time. In most cases, the cavern is used to store hydrocarbons (crude oil, LPG or natural gas; see Thoms and Gehle, 2000). Storing oil-field wastes in salt caverns has been considered by several authors (e.g., Rolfs et al., 1996, and Tomasko et al., 1997), and several such storages are being operated in Texas. An abundant literature is available on all aspects of solution-mining techniques, including safety. The Solution Mining Research Institute (SMRI), which represents companies, consultants and research centers involved in the

solution-mining industry, has published hundreds of technical papers dedicated to these questions.

Here, in order to be specific the authors assume that 600-m^3 of tritiated water must be disposed of each year for 20 years. We also assume that a cavern with a volume of 16,000 m^3 is created at a depth of approximately 1000 m below ground level. The proposed depth is purely indicative, but a significantly larger depth would generate large creep rates, as will be explained below.

In our operating scenario, the liquid wastes (tritiated brines) occupy the cavern bottom, and the upper part of the cavern is filled with a lighter fluid (e.g., fuel oil). The amount of stored liquid waste will increase with time, and an equivalent volume of fuel oil will be progressively withdrawn from the cavern so that the total (brine + hydrocarbon) volume is kept constant. There are no physical or chemical exchanges between the two liquids (except for a small amount of gas generated at the interface due to radiolytic effects), so radioactivity is confined to the lower part of the cavern. An alternative solution consists in filling the upper part of the cavern with air, although this solution may imply severe cavern convergence, which will be discussed later.

The well is equipped with a steel casing whose shoe is anchored above the cavern neck. One or two strings are set in the well, delimiting one or two annular spaces and a central volume (see Figure 1). Liquid waste is injected in the cavern through the central tubing, and fuel is removed through the annular space. When the system is at rest, the well's central tubing and annular spaces are filled with fuel oil, and the well-head pressure is null or small. This results in a cavern pressure that is smaller than the halmostatic pressure (obtained when the well is filled with brine) — and even smaller than the hydrostatic pressure (obtained when the well is filled with soft water).

CAVERN TIGHTNESS

In the proposed storage method, waste confinement in a salt cavern is based on:

(1) the extremely small permeability of rock salt; and

(2) the depressurization of the cavern, which has a pressure smaller than the natural pore pressure of the brine in the rock mass.

Salt Permeability

Rock salt exhibits a very low permeability, because the hydraulic conductivity of its matrix is extremely small and because no fractures exist in a massive salt formation (except, perhaps, in some disturbed zones encountered at the fringe of salt domes). Figures as small as K = 10^{-22} m^2 to 10^{-20} m^2 are reported. Several authors even believe that most of this (small) permeability is induced by the cavern creation and operation (more precisely, by tensile or high deviatoric stresses developed at the cavern wall when cavern fluid pressure is very high or very small, respectively). In fact, few reliable in-situ test results are available, as permeability is so small that its measurement is beyond the standard techniques used for more

permeable rocks (say, rocks with permeability larger than $K = 10^{-17}$ m^2). For example, experiments performed in an air-intake shaft at the WIPP site provide permeabilities as low as $K = 10^{-21}$ m^2 for undisturbed salt (Dale and Hurtado, 1997). In the Etrez upper salt formation in France, where anhydrite and clay interbeds are present, a one-year test in a 1000-m deep well gave $K = 6 \times 10^{-20}$ m^2 (Durup, 1994). More recently, at the same site, an 18-month long test in a full-sized cavern provided $K = 2 \times 10^{-19}$ m^2 (Bérest et al., 2001a.), which is consistent with the generally accepted effects of scale on rock permeability (Brace, 1980).
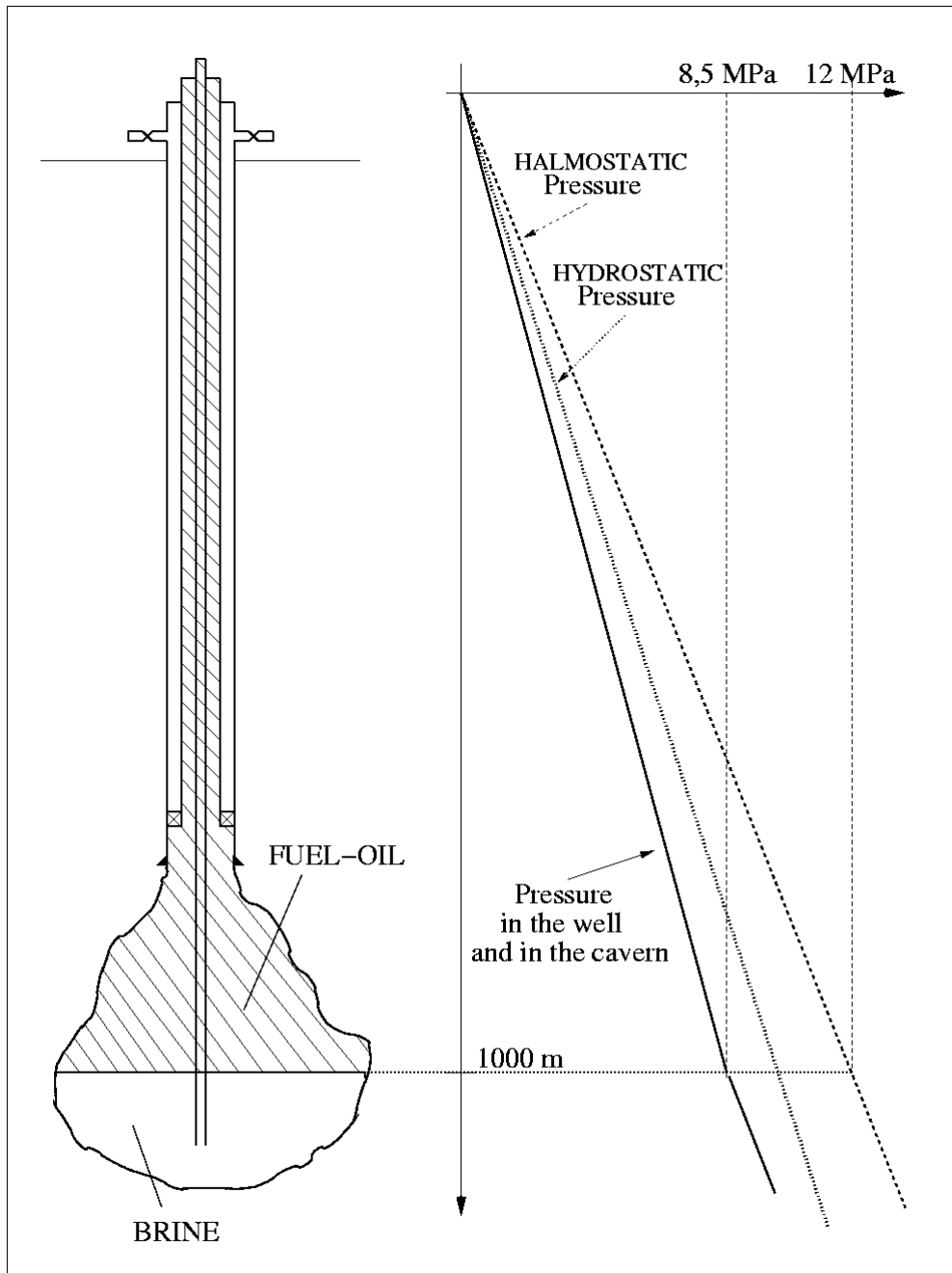

Figure 1   Pressure distribution in the well. Cavern fluid pressure is smaller than rock mass pore pressure which is assumed to be close to halmostatic pressure, resulting in a (tiny) inward brine flow.

Pore Pressure

Natural brine pressure in the interconnected pores or cracks cannot be measured directly. Durup's test, mentioned above, proved that a linear relation did exist between the brine- seepage rate and the pressure build-up in the Etrez well. Extrapolating this relation proves that no flow takes place when cavern pressure is equal to halmostatic pressure, which strongly suggests that the natural pore pressure is equal to the halmostatic pressure. Even if no clear physical argument can be invoked, this hypothesis seems to be widely accepted. The distribution of pore pressure in the formation is, then,

$$P_o \text{ (MPa)} = 0.012 \, H \text{ (m)} \qquad (1)$$

where H is the cavity depth.

However, a test performed at the WIPP mine excavated in the Salado formation provided higher figures (P_o = 0.014 H; see Howarth et al., 1991). Considering the halmostatic distribution expressed by (1), this formula indicates that no brine flow to or from a cavern takes place if the cavity well is filled with saturated brine and opened to atmosphere when, in this instance, cavern brine pressure (P_i) is equal to natural pore pressure (P_o).

Fluid Seepage

How small salt permeability is can be illustrated by a simple example in which, as a rough estimation, $K = 10^{-20}$ m^2 and there is a 3-MPa excess of pressure in a storage cavern (compared to the natural pore pressure in the rock mass; in other words, $P_i - P_o = 3$ MPa). For a spherical cavern of radius R, the steady-state flow from the cavern is

$$\frac{Q}{V} = 3\frac{K}{\eta}\left(\frac{P_i - P_0}{R^2}\right) \qquad (2)$$

where η is the fluid viscosity ($\eta = 1.2 \times 10^{-3}$ Pa.s for saturated brine) or Q/V = 10^{-4} per year in a 16,000-m^3 cavern or Q = 1.6 m^3/year, a very small seepage rate. Of course, this (small) flow is directed outward, because the cavern pressure is larger than the natural pore pressure in standard hydrocarbon-storage conditions.

The storage principle proposed in this paper consists of setting the cavity pressure (P_i) smaller than the natural pore pressure (P_o) to promote an inward flux of brine from the rock mass to the cavity and preventing any fluid seepage. This idea has been used for years with remarkable success in the context of LPG storage in unlined caverns at shallow depth (see, for example, Bérest, 1989).

A (relatively) small and constant cavity pressure (P_i) can be obtained by filling the well and the cavern top with a light fluid (e.g., fuel oil, which has a density of 850 kg/m^3), resulting in a cavern pressure, P_i, of

$$P_i \text{ (MPa)} = 0.0085\, H \text{ (m)} \qquad\qquad (3)$$

This small brine flow is directed toward the cavern — i.e., 1 m^3 to several m^3 per year toward a 16,000-m^3 cavern if we assume the same cavern shape and depth, and the same rock or fluid properties as before. Flow from the cavern to the rock mass cannot exist.

The major drawback of this system is that cavity pressure is smaller than in most hydrocarbon-storage caverns, which results in more severe convergence rates. This aspect will be discussed later.

SAFETY ISSUES

Well Integrity

Because the cavern pressure is below the hydrostatic pressure, one crucial issue with regard to storage "tightness" is the integrity of the well — not salt permeability. The casing, cementation and casing shoe are, potentially, the weakest points.

Loss of casing tightness is known to have occurred in hydrocarbon storages, which have much higher fluid pressure. A discussion of factors contributing to casing leaks and the description of such loss can be found in Bérest et al. (2001b.). Such occurrences are extremely unlikely in double-cased wells; well tightness can be periodically checked through mechanical integrity tests (Crotogino, 1994). Furthermore, in the suggested concept, the liquid wastes are confined to the cavern bottom, so any failure in well integrity will lead to a fuel-oil leak — not a waste leak.

Waste Handling

Tritiated water is highly corrosive. Any failure of the surface equipment (pumps, well-head, tubes) that causes water spills can induce harmful consequences for the crew and the environment. The probability of such a failure must be as close to zero as possible. The simplest waste-handling system is the best. A hydrocarbon-filled well allows the flow of liquid wastes, which are heavier than hydrocarbons, to be purely gravity-driven. Thus, pumps are unnecessary.

Retrievability

It is always possible technically and economically feasible to pump out a high percentage of the stored waste from a cavern through the central injection tube which can be lowered to cavity bottom.

Human Intrusion

The period of time during which wastes must be confined is relatively small (100 to 200 years). For such short time periods, the risk of inadvertent human intrusion is not

considerable. Events such as willful damage, plane crashes or earthquakes can lead to well-head failure. The stored liquid, being heavier than hydrocarbons and soft water, will remain at the cavern bottom, with no risk of upward migration to aquifer layers or ground level. Furthermore, the harmful liquid (saturated brine) is not drinkable unless it is diluted in a very large volume of soft water.

CAVERN CREEP

The Influence of Depth

For deep caverns, creep can be a concern. Some case studies (Boucly and Legreneur, 1980; Baar, 1977; Kuhne et al., 1973) describe a few deep caverns that have experienced large shrinkage, or typical volume loss rates ranging from 3% to 30% per year. Large volume losses occur in deep natural gas caverns for two reasons:

(1) high geostatic pressure due to the great depth; and

(2) low internal pressure, especially when the cavern is almost empty.

In other words, the critical parameter is not cavern depth — i.e., the geostatic pressure (P_∞), but rather the gap ($P_\infty - P_i$) between the geostatic pressure at cavern depth and the cavern fluid pressure. A natural gas cavity at medium depth can creep as quickly as a brine-filled cavity at greater depth.

In order to clarify this statement, we estimate that the geostatic pressure (P_∞, in MPa) due to the overburden weight at depth H (in meters) is

$$P_\infty \,(\mathrm{MPa}) = 0.022\, H \,(\mathrm{m}) \tag{4}$$

This results from an average rock density of 2200 kg/m^3.

If the well is filled with a saturated brine with a density of 1200 kg/m^3, and if no additional well-head pressure is applied to the brine column, the internal pressure in a cavern located at depth H is equal to the halmostatic pressure ($P_i = P_0$; see (3)), and the gap between the geostatic pressure and the internal pressure is simply proportional to depth:

$$P_\infty - P_i \,(\mathrm{MPa}) = 0.01\, H \,(\mathrm{m}) \tag{5}$$

For example, at a depth of 1000 m, the gap is 10 MPa.

Fuel-oil density (850 kg/m^3) is lower than brine density (1200 kg/m^3). When substituting fuel oil for brine in the well, the fluid pressure in the cavern itself is lowered significantly, resulting in faster creep. This effect must be thoroughly evaluated — too-high a convergence rate can be troublesome, because it reduces the available cavern volume.

When the well of a cavern located at depth H is filled with fuel oil, the well-head pressure in the cavern is

$$P_i \text{ (MPa)} = 0.0085\, H \text{ (m)} \tag{6}$$

This means that the gap between the geostatic and internal pressure is

$$P_\infty - P_i \text{ (MPa)} = 0.0135\, H \text{ (m)} \tag{7}$$

The difference for the case of a brine-filled well will be discussed in the next section.

The Mechanical Behavior of Salt

The mechanical behavior of salt exhibits a fascinating complexity, and several aspects of it are still open of discussion. However, experts agree on the following features of importance to the problem under discussion.

1. Salt behavior is elastic-ductile when short-term compression tests are considered, but, in the long term, salt behaves as a fluid in the sense that it flows even under very small deviatoric stresses.

2. Creep rate is a highly non-linear function of applied deviatoric stress and test temperature.

Furthermore, experts generally distinguish between:

(a) steady-state (or secondary) creep (Steady-state creep is reached after some time (several weeks) when a constant mechanical loading is applied to a rock sample; it is characterized by a constant creep rate that is a function of the (constant) temperature and stress applied during a test.); and

(b) transient (or primary) creep. (Transient creep is triggered when the stress applied to a sample is changed suddenly. It is characterized by high initial rates (following a load increase) that slowly reduce to reach a steady state.)

A similar distinction between steady-state and transient behavior in a salt cavern can be made when, instead of a sample axial-strain rate ($\dot\varepsilon$), we consider the volumetric strain rate of the cavern ($\dot V / V$) and, instead of the uniaxial stress applied on the sample, we consider the difference ($P_\infty - P_i$) between the natural geostatic pressure (P_∞) at cavern depth and the internal brine pressure in the cavern (P_i). However, this analogy must be corrected slightly. The transient mechanical effects in a cavern last much longer than in a rock sample: stress distribution is not uniform in the vicinity of a cavity, in contrast to the situation that prevails during a uniaxial test on a cylindrical rock sample, and the stress change due to the change in brine pressure must propagate throughout the rock mass before steady-state behavior is reached. In other words, transient cavern behavior results from the transient rheological behavior of salt combined with a factor of geometrical origin.

Returning to the uniaxial behavior of a rock sample, the steady-state creep behavior is described by the following expression, sometimes called the Norton-Hoff (or power-) law:

$$\dot{\varepsilon} = A \ \exp(-Q/\Re T) \ \sigma^n \tag{8}$$

where A, n and $-Q/\Re$ are parameters, σ is the uniaxial applied stress, and T is the (absolute) test temperature. Exponent n ranges from 3 to 6, and $Q/\Re$ ranges from 4000 K to 10,000 K.

Typical values provided by Van Sambeek (1993) [who compiled data from De Vries (1988) and Minson et al. (1989)], Senseny (1984), Wawersik (1984) and Pouya (1991) are displayed in Table 1.

Table 1 Parameters for different salts

Facility	n	Q/R (K)	A (year$^{-1}\times$MPa^{-n})	Sphere/Cylinder Ratio
Avery Island (after D.V)	3.14	6495	1.30×10^4	1.81
WIPP	5.0	5035	1.04	2.37
Etrez	3.1	4100	0.64	1.80
Avery Island (after S. and al.)	4.0	6565	2081	2.05
Salina	4.1	8715	2.7752×10^5	2.08
Palo Duro – Unit 4	5.6	9760	1.806×10^5	2.58
Palo Duro – Unit 5	5.3	9810	2.52×10^5	2.47
Salado (WIPP7)	5.09	8333	3.67×10^4	2.40
Asse (BGRC1)	6.25	9969	2.51×10^4	2.84
West Hackberry (WH1)	4.73	6606	452.31	2.28
West Hackberry (WH2)	4.99	10766	0.94	2.37
Bryan Mound (BM3C)	4.54	7623	1.32×10^3	2.22
Bryan Mound (BM4C)	5.18	8977	1.04×10^5	2.43
Bayou Choctaw (BC1)	4.06	5956	64.03	2.07

It should be noted that these parameters are not recommended by the authors, who, in some cases, suggest a more elaborate constitutive equation. In addition, the authors do not use the same units, and we have converted the constants to get years and Megapascals, which are more convenient for the problem being discussed.

Direct comparison of data is difficult: when A is large, $\exp(-Q/\Re T)$ is small, resulting in less-scattered strain rates, except for Avery Island salt (Van Sambeek, 1993). It is convenient to compare these data by computing how a spherical cavern behaves as a function of depth.

For this purpose, the uniaxial expression (8) must be generalized in a 3D formulation:

$$\dot{\varepsilon} = A \ \exp \ (-Q/\Re T) \ \frac{1}{n \ + \ 1} \ \frac{\partial \ (\sqrt{3J_2})^{n+1}}{\partial \underline{\underline{\sigma}}} \tag{9}$$

where $J_2 = \dfrac{1}{2} S_{ij} \ S_{ji}, S_{ij} = \sigma_{ij} - \dfrac{1}{3}(\sigma_{ii})\,\delta_{ij}$.

Using this formulation, a closed-form solution can be obtained for the idealized case of a perfectly spherical (or cylindrical) cavern that, over a long period of time, is subjected to an internal pressure (P_i) smaller than the natural geostatic pressure (P_∞) at cavern depth.

For a spherical cavern, the steady-state relation for volume rate change, temperature and pressure is as follows:

$$\frac{\dot{V}}{V} = -\frac{3}{2}\left[\frac{3}{2n}(P_\infty - P_i)\right]^n A \exp\left(-\frac{Q}{\Re T}\right) \qquad (10)$$

A similar formula for a cylindrical cavern has been given by Van Sambeek (1993):

$$\frac{\dot{V}}{V} = -\sqrt{3}\left[\frac{\sqrt{3}}{n}(P_\infty - P_i)\right]^n A \exp\left(-\frac{Q}{\Re T}\right) \qquad (11)$$

Hence, the cylindrical case can be deduced from the spherical case by multiplying the latter formula by $(2\sqrt{3}/3)^{n+1}$. Among all possible shapes, the slowest volume-loss rate is reached when the cavern is spherical.

Real caverns are neither spherical nor perfectly cylindrical. A flat cavern (i.e., one having a diameter much larger than its height) behaves fairly distinctly, but the spherical/cylindrical cases provide a good illustration of the behavior of most regular-shaped caverns.

One immediate consequence of the above-mentioned feature (a), which is captured by (10) and (11), is that, as long as the cavern fluid pressure is smaller than the geostatic pressure, the cavern shrinks.

Cavern convergence rate is a highly non-linear function of cavern depth, as both geothermal temperature (T) and geostatic pressure (P_∞) are an increasing function of cavern depth. To perform computations, the following two assumptions have been made:

1. The temperature at depth is given by T (K) = 288+0.03 H (m). In other words, the surface temperature is 15°C; at a depth of 1000 m, the rock temperature is 45°C.

2. For the case of a well filled with fuel oil,

$$P_\infty - P_i \text{ (MPa)} = 0.0135 \text{ H (m)} \qquad (12)$$

and for the case of a brine-filled well,

$$P_\infty - P_i \text{ (MPa)} = 0.01 \text{ H (m)} \qquad (13)$$

The results are given on Figures 2 and 3, which display cavern volume-loss rate as a function of cavern depth. The case for a cylindrical cavern can easily be obtained by multiplying the volume loss rate by the ratio given in Table 1. The volume loss rate is larger in the case of a well filled with fuel oil, as expected. Depths larger than 1000 m must be avoided. A typical value, obtained in the case of a 1000-m deep cavern leached out from Etrez salt formation is 3×10^{-4} year^{-1} (brine-filled well) or 7.4×10^{-4} year^{-1} (well filled with fuel oil). The former figure is very close to the figure observed during an in-situ test (Brouard, 1998); the latter figure implies that total convergence after 200 years would be 15%, an admissible figure provided that the volume of stored tritiated water is smaller that cavern volume (say, one-half), which allows some cavern volume shrinkage without inconvenience. The convergence rate would be more severe if air (instead of fuel oil) were used as a blanket fluid. However, air pressurization and/or shallower cavern depth allow the severity of this problem to be mitigated.

CONCLUSION

Storing hazardous waste in salt caverns is an appealing solution when the toxicity of the waste vanishes after a few decades or centuries. A suitable selection of cavern fluid pressure prevents too fast a convergence rate and allows a small amount of rock-mass fluid flow to be directed toward the cavern, preventing any fluid seepage from the cavern.
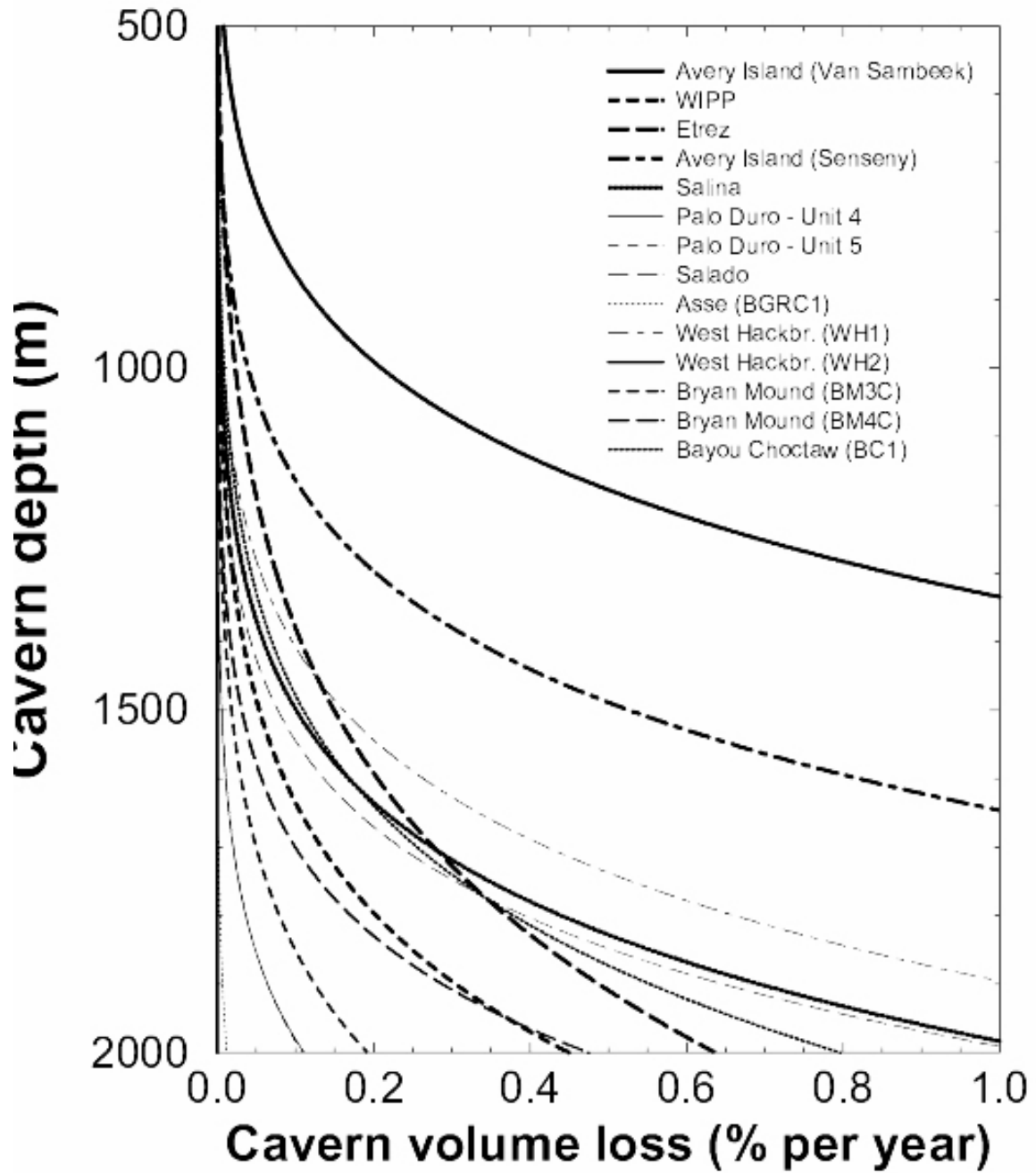
Figure 2. Yearly cavern volume loss is extremely small when cavern depth is smaller than 1000 meters.
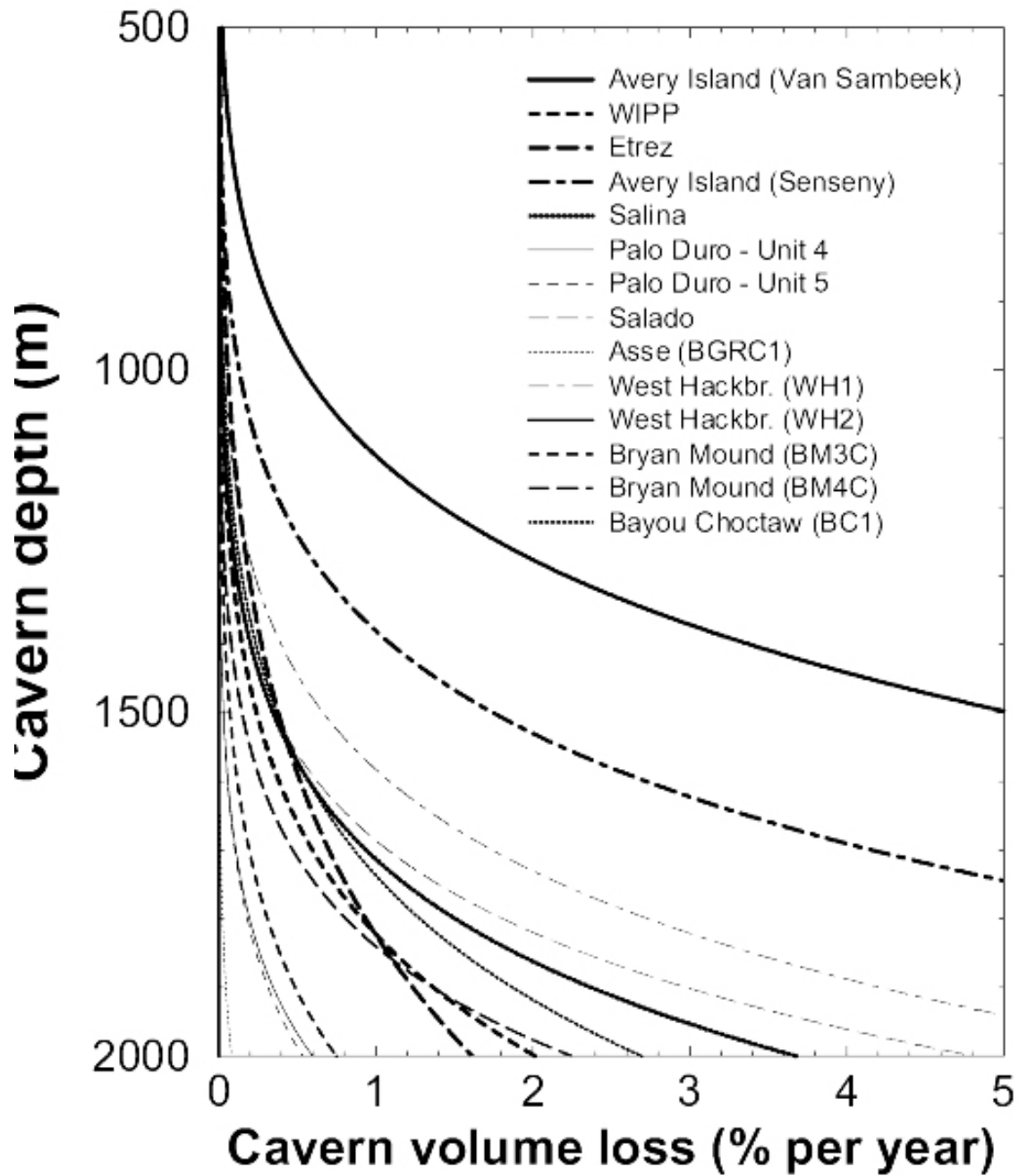
FUEL OIL FILLED WELL

Figure 3. Yearly cavern volume loss is significantly larger than in the case of a brine-filled well, but still admissible.

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