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L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

THE LEACHING OF RADIOACTIVITY FROM HIGHLY RADIOACTIVE GLASS BLOCKS BURIED BELOW THE WATER TABLE: FIFTEEN YEARS OF RESULTS

by

W.F. MERRITT

Presented at IAEA/NEA International Symposium on the Management of Radioactive Wastes from the Nuclear Fuel Cycle, 22-26 March, 1976, Vienna, Austria

Chalk River Nuclear Laboratories
Chalk River, Ontario

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Lixiviation de la radioactivité provenant de blocs de verre fortement radioactifs enfouis sous la nappe aquifère: quinze années de résultats

par

W.F. Merritt

Présenté au colloque international IAEA/NEA concernant la gestion des déchets radioactifs provenant du cycle de combustible nucléaire, Vienne, Autriche, 22-26 Mars 1976.

Résumé

Les résultats provenant de deux enfouissements expérimentaux de produits de fission à haute activité, incorporés dans du verre à base de syénite néphénilique, montrent que les déchets nucléaires provenant du traitement du combustible pour centrales nucléaires totalisant 30 000 MWe pourraient être incorporés dans de tels récipients de verre et stockés sous la nappe aquifère dans la zone de gestion des déchets des Laboratoires Nucléaires de Chalk River, sans effets nocifs pour l'environnement.

L'Energie Atomique du Canada, Limitée
Laboratoires Nucléaires de Chalk River
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ABSTRACT

The results from two test burials of high-level fission products incorporated into nepheline syenite glass indicate that the nuclear wastes from fuel processing for a 30,000 MWe nuclear power industry could be incorporated into such glass and stored beneath the water table in the waste management area of Chalk River Nuclear Laboratories (CRNL) without harm to the environment.

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1. INTRODUCTION

One of the problems associated with the nuclear power industry is the ultimate storage of the radioactive wastes produced. The processing of spent fuels results in the accumulation of highly radioactive solutions which must be stored in such a manner that the chance of release to the environment is minimal. As yet, this is not a serious problem in Canada, since the CANDU* heavy water reactor system makes such efficient use of the uranium fuel that processing is not as yet economic. At present, spent fuel is stored under water at the reactor sites. Future storage methods will be discussed at this symposium by Mayman, Scott and Barnes. However, it is envisioned that in the future it will become economically feasible, if not mandatory, to process spent fuel to recover the fertile and fissionable isotopes from it. The need is more acute if one is operating or intends to operate reactor systems using enriched fuel.

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*Canada Deuterium Uranium

At present, fuel processing wastes are still generally stored as concentrated solutions in special tanks in a controlled area. Elaborate precautions must be taken to prevent or contain leakage, and monitoring plus maintenance must be provided. Because of the mobility of liquids there is general agreement that eventual solidification of these wastes will be required, either in the tanks or as highly resistant glasses or ceramics [1], with storage in specially engineered facilities such as vaults or salt mines.

Many processes have been studied for converting highly radioactive solutions into solids [2]. The ultimate in such an approach to the problem would be to develop a material, the leaching rate of which would be so low that it would be possible to store it by burial in the ground without containment. The development of such a material at Chalk River Nuclear Laboratories (CRNL) has been described by Watson, Aikin and Bancroft [3]. The material is a glass based on nepheline syenite, a naturally occurring alumino-silicate mineral. To prepare the glass, a mixture of 85% nepheline syenite and 15% lime was combined with a nitric acid solution of fission products in ceramic crucibles. The resulting gel was dried, denitrated at 900°C and then melted at 1350°C. Volatile components, mainly ruthenium and cesium were adsorbed on a heated bed of fire brick and iron oxide.

By 1960, laboratory operations had produced glass blocks sufficient for two test burials. The first of these has been described by Bancroft and Gamble [4]. Twenty-five blocks containing about 300 Ci of aged fission products and made to a highly resistant formula were buried in a 5 x 5 block grid below the water table in the CRNL waste management area in August 1958. When early results from this test showed leaching rates too low to be readily measurable, a second test was carried out and has been described by Bancroft [5]. Twenty-five blocks containing about 1100 Ci of aged fission products were buried in a tighter 5 x 5 block grid in a similar location in May 1960. Laboratory leaching tests of this glass, which contained a higher proportion of uranium and corrosion products showed a leaching rate about ten times higher than that found for glass used in the first test. Considering this higher dissolution rate, larger quantity of fission products

and tighter configuration, it was calculated that the amount of radioactivity leaving the blocks should be higher by a factor of 150 than the first disposal.

A description of the methods used in monitoring the second disposal has been given by Merritt and Parsons [6]. The ground water velocity past the blocks was measured using tritium as a tracer. The ground water downstream from the blocks has been sampled regularly and analyzed radiochemically. In 1963, 1966 [7] and 1971, soil samples were taken from downstream of the blocks and the patterns and concentrations of radioactivity which had left the blocks was determined. Results from this test have now been obtained over a period of 15 years.

2. RESULTS

Three ground water samplers positioned 0.6 m apart and 1 m downstream of the glass blocks have been sampled monthly from April to November each year since 1960. The water was analyzed for ^{90}Sr and the average content for each year is shown in Table I. The ground water velocity was measured to be 18 cm/d, the flow cross section of the blocks was 5600 cm^2 and the soil porosity was 38%. Therefore 39 ℓ/d of water flows past the blocks. Using this figure the amount of ^{90}Sr leaving the blocks per year was calculated and is shown in the same Table.

The amount of ^{90}Sr leached from the blocks has remained essentially constant for the past seven years. The amount of ^{90}Sr that has left the blocks in 15 years is about 1.5 mCi, 90% of which was in the first two years.

Sampling in 1971 showed that the ^{90}Sr front had reached 33 m from the glass blocks. This is in good agreement with predictions from the surveys in 1963 and 1966, and indicates that ground water conditions have not altered significantly since the start of the experiment.

Laboratory studies of the glass used in the first test indicated that it would have a leaching rate about 1/10 of that

TABLE I. STRONTIUM-90 LEAVING SECOND TEST DISPOSAL

Year	^{90}Sr Concentration in Ground Water μCi/ℓ	Total ^{90}Sr Leaving Blocks Per Year μCi (corr. to 1960)	% of 15 a Total
1960	1.3×10^{-1}	1060	70
1961	2.4×10^{-2}	350	23
1962	1.0×10^{-3}	15	1.0
1963	1.4×10^{-3}	21	1.4
1964	1.0×10^{-3}	15	1.0
1965	7.0×10^{-4}	11	0.73
1966	2.6×10^{-4}	4.2	0.28
1967	2.1×10^{-4}	3.5	0.23
1968	1.7×10^{-4}	2.9	0.19
1969	1.9×10^{-4}	3.3	0.22
1970	1.7×10^{-4}	3.0	0.20
1971	1.3×10^{-4}	2.3	0.15
1972	2.0×10^{-4}	3.8	0.25
1973	1.2×10^{-4}	2.3	0.15
1974	1.2×10^{-4}	2.5	0.17
		1500 μCi in 15 a	

used in the second test. To date, it has not been possible to detect any ^{90}Sr in the water samples taken downstream of the first test disposal. The sensitivity of our sampling and counting methods was such that it should have been possible to detect ^{90}Sr in the ground water if the leaching rate was 1/5 that of the glass used in the second test. Therefore, we can assume that it is possible to make glass with a leaching rate five times lower than that observed for the second test.

3. DISCUSSION

Thirty-nine ℓ/d of water flows by the blocks. The concentration of ^{90}Sr in the glass blocks was 1.0×10^{-2} Ci/g. The total surface area of the blocks is 1.1×10^{4} cm^2. Therefore, using the values for ^{90}Sr in the ground water given in Table I, it is possible to calculate the amount of glass being leached daily. For example in 1974,

$$\frac{1.2 \times 10^{-4} \times 39}{1.0 \times 10^{-2} \times 10^{6} \times 1.1 \times 10^{4}} = 4 \times 10^{-11} \text{ g cm}^{-2} \text{ d}^{-1}.$$

For the past seven years the ^{90}Sr leaving the blocks has remained essentially constant and averaged 1.6×10^{-4} μCi/ℓ. Table II shows the leaching rate based on the ^{90}Sr, up to the end of 1974.

In a study made in 1958, Watson, et al. [8] calculated the parameters needed to evaluate methods of permanent storage. Using their data as applied to a 1200 MWe CANDU heavy water reactor, and assuming that all the fuel is processed and the fission products are incorporated in nepheline syenite glass and buried below the water table in the CRNL waste management area, then it is possible to use the leaching rates shown in Table II and calculate the cumulative release of ^{90}Sr from the blocks. The results are shown in Figure 1. After 200 years of continual operation of a 1200 MWe reactor, about 200 Ci of ^{90}Sr would be in the plume downstream of the blocks. An examination of the curve shows that about 4 Ci/a of ^{90}Sr leaves the blocks. After about 30 years the decay of ^{90}Sr begins to

TABLE II. LEACHING RATE OF GLASS BLOCKS

Year	Leaching Rate $g\ cm^{-2}\ d^{-1}$
1960	4×10^{-8}
1961	7×10^{-9}
1962	4×10^{-10}
1963	3×10^{-10}
1964	3×10^{-10}
1965	2×10^{-10}
1966	8×10^{-11}
1967	6×10^{-11}
1968-74	5×10^{-11}

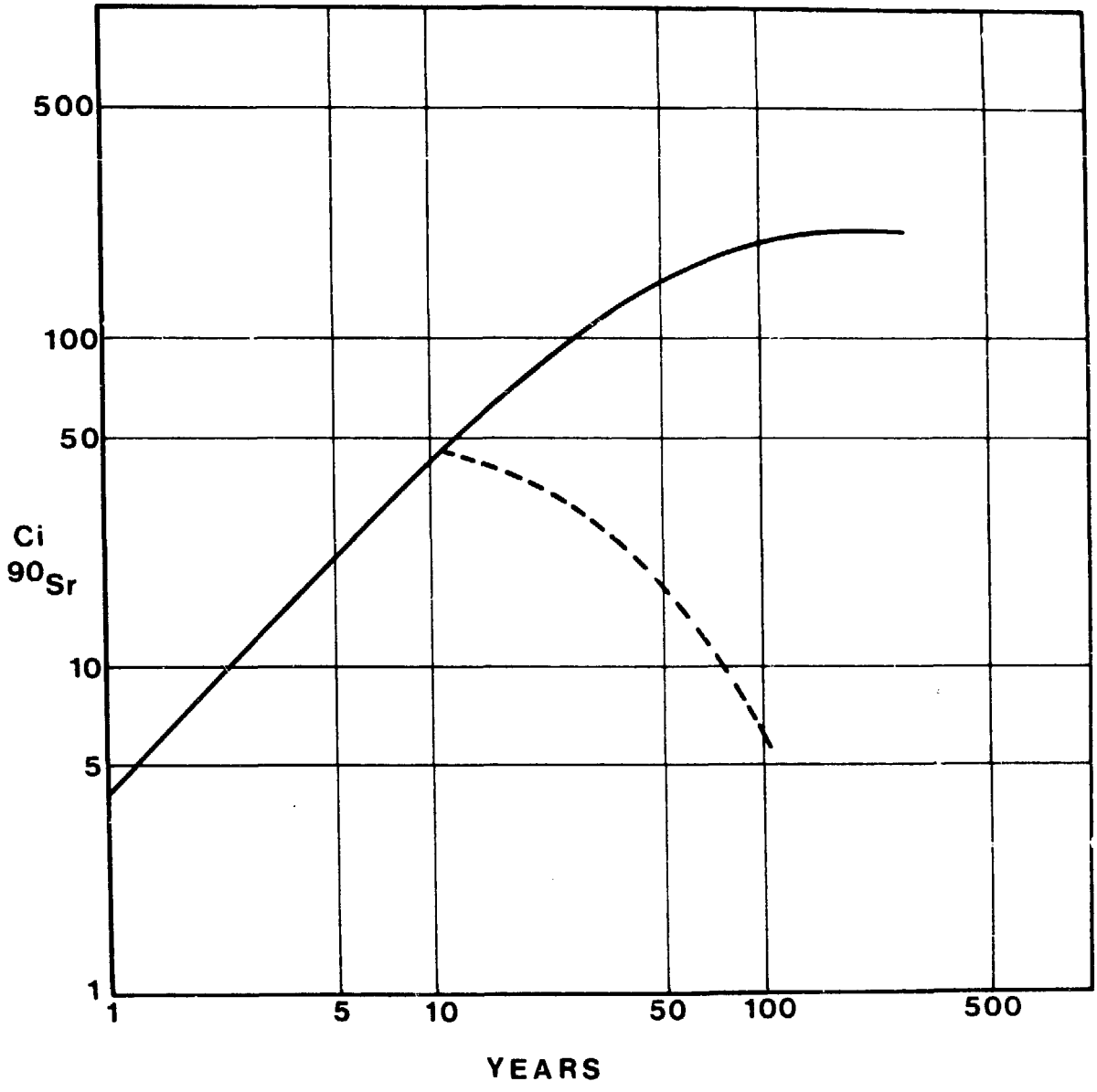

FIG. 1. Total ^{90}Sr leached from blocks for 1200 MWe CANDU reactor.

become apparent and equilibrium is reached in about 200 years. The lower curve shows the result of stopping operations after ten years when a maximum amount of 40 Ci would be leached.

Suppose such a waste storage facility was set up at CRNL and the glass blocks buried in the Perch Lake basin. The measured velocity of ^{90}Sr from the second test disposal has been 33 m in 12 a or 2.75 m/a. The distance from a suitable storage site to Perch Lake is 500 m. Therefore it would take 180 years, or six half-lives for the ^{90}Sr to reach the lake. By this time, the 4 Ci/a released from the blocks would have decayed to

$$\frac{4 \times 10^3}{64} = 62.5 \text{ mCi/a} \quad \text{or } 170 \ \mu\text{Ci/d}.$$

High-level wastes from a fuel processing plant servicing a 30,000 MWe nuclear industry would, under these conditions, release only 5 mCi/d to Perch Lake.

Methods of improving the performance of the glass blocks are available. For example, the first test demonstrated it was feasible to manufacture glass with a leaching rate at least five times lower than the glass used in the second test. Also, the data in Table I shows that over 90% of the ^{90}Sr leaving the blocks did so in the first two years. Laboratory tests [3] have shown that pre-leaching of the blocks would prevent this original large release. The pre-leaching might have to be done *in situ*, and would generate a secondary liquid waste stream but would seem to be quite feasible. Again, making larger blocks with a lower surface to volume ratio than the blocks used would reduce the leaching rate.

It should be pointed out that these figures only hold if the glass is actually buried beneath the water table and in a configuration such that the ground water temperature is not significantly increased. Laboratory results [3] have shown that high water temperatures increase the rate of leaching considerably. Also it has been shown that devitrification of the glass will also increase the leaching rate. On the other

hand, tests have shown that the irradiation received by the glass has no effect on the leaching rate.

When the glass was buried for the second test, three of the blocks lodged above the grid and have not been considered in the calculations. It should be possible to recover at least one of these blocks without disturbing the experiment. It would then be feasible to examine and test the block in a hot cell. The author would be interested to get the opinion and advice of others working in this field on the usefulness of such a procedure and on the type of examination and measurements required.

4. CONCLUSION

The results indicate that the incorporation of high-level nuclear wastes in nepheline syenite glass and burial of the glass directly in the soil in the CRNL controlled area would be a suitable method for permanent storage of such wastes from a 30,000 MWe nuclear power industry.

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