Operation "Hot Mole"

Self-burial: an elegant solution to the problem of storing high-activity radioactive effluents?

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Scale of the radioactive waste storage problem

The storage of radioactive residues from nuclear power plants is a crucial problem, both because of the rapid increase in production levels forecast for the coming years and because of the health and ecological implications of the question, and is one to which a solution must be found in the near future which will afford maximum security in matters of safety and protection.

Table I shows the importance which the problem of the disposal of waste from power reactors is likely to assume in the Community. The data on the production and activity of high-activity waste were taken from an article which appeared recently in this review (1). They are based on the assumption of a minimum nuclear energy development programme in the Six (40 000 MWe installed in 1980, 400 000 MWe in 2000).

On the basis of data supplied in a recent American study (2), we have estimated the thermal power generated by all high-activity products accumulated by the end of every decade.

In this evaluation, it has been assumed that the waste is stored for one year after the irradiated fuel is unloaded and that the reprocessing operations dilute the pure fission products by a factor of 10. The results are shown in the last line of Table I. It will thus be noted that the power generated by the total waste accumulated by the year 2000 will be of the order of 700 MW.

This value, which relates to only a minimum nuclear energy development programme in the Community, represents the power generated by a fair-sized reactor. It is also interesting to ascertain the upper limit to the production and activity of solid waste.

The total energy consumed to date in the Community is of the order of 900 million tce (1 tce = 0.36 MWd). Let us imagine that this energy is at present produced exclusively by nuclear means and that consumption increases by 5% a year. On the assumption that the efficiency of the power plants producing the energy vector (5) is 50% and that 10 000 MWd produce 25 litres of solidified waste (2), by 2000 more than 110 000 m^3 of waste will have accumulated, generating something like 15 000 MW.

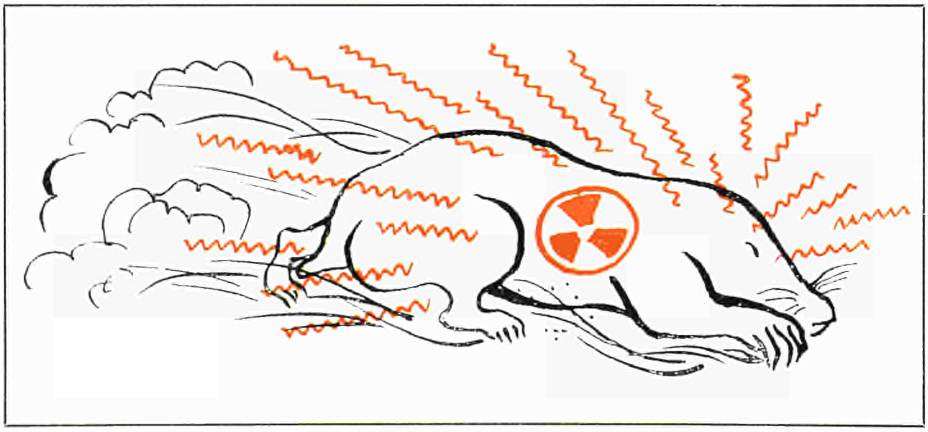
These figures show clearly that the problem of heat removal from nuclear waste will, in a few decades, be extremely grave. These considerations quite naturally prompt the question as to whether a storage technique is conceivable in which this heat could be used to bury waste in the lower strata of the subsoil. Such a solution is proposed in this article. Before examining it in detail, we propose to review briefly the various storage techniques currently in use.

Techniques used for storing high-activity waste [(2), (3)]

There is to date no long-term, large-scale industrial practice for the processing of irradiated fuel from power reactors. The disposal of high-activity effluents, in particular, is only just beginning to be a matter of concern to reprocessing plants.

Storage in liquid form has been normal practice for some 20 years; the effluents, in the form of acid solutions, are contained in stainless steel tanks or, if neutralized, in carbon steel tanks. The heat generated by radioactive decay is usually sampled during storage; this is done by immersing coils containing water in the active solution. Around 350 000 m^3 of waste are currently stored in the United States in more than 200 underground tanks.

Storage in liquid form by no means fulfils all the safety conditions necessary,



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* With acknowledgements to Mr S. GIULIANI and Mr L. KESTEMONT for their assistance.

since some 15 carbon steel tanks have so far been known to develop leaks, causing contamination of the soil. Moreover, this type of storage takes up a great deal of space and will not be practicable when the quantities of effluent are much higher than at present.

Laboratories in a number of countries are thus considering the development of other types of radioactive waste storage. To date, storage in solid form seems to be the only technique offering an appreciable reduction in volume and an increase in safety.

Four high-activity liquid effluent solidification processes have been developed and tested in pilot installations during recent years (2). They are: calcination in crucibles, spray solidification, phosphated glass encapsulation and fluidized-bed calcination. Each of these techniques requires a supply of heat to rise the temperature and remove the volatile constituents (water, nitrates). The solid left after cooling is relatively stable chemically.

The American regulations tend to require compulsory solidification of fission products in the five years after fuel unloading and storage in a "graveyard" (salt mine) in the 10 years following discharge. A different philosophy is beginning to emerge which recommends immediate solidification on leaving the reprocessing plant and "cooling" in solid form on site for three to five years.

In these cases, the process used would probably be calcination of effluents with an inert diluent (e.g. aluminium nitrate). The cooled solid could be incorporated in a less leachable medium (such as glass or metal) before being sent to the graveyard.

In Europe no regulations or draft regulations have so far been proposed. It should also be mentioned that studies are currently in progress in the United States to investigate the possibility of dumping the most active waste on other planets or the sun.

The principle of self-burial

As we have seen, the dissipation of heat in effluents from irradiated fuel reprocessing installations is an important problem in the usual storage techniques.

The self-burial technique which we suggest in this article, on the other hand, consists of using the heat generated by radioactive decay to dump the waste in the deep strata of the subsoil. It is possible to pack high-activity solid waste in suitable containers and bury these in the ground. If the heat generated by the products is higher than a critical value determined subsequently, after a certain period of time the soil in contact with the container will melt and a further burial process, this time automatic, will take place. Provided that the container is denser than the soil and capable of withstanding the melting point of the soil, the self-burial process may be prolonged for as long as the power source, in the form of active waste, can supply the necessary heat.

This, therefore, is a method of burying nuclear waste in the deeper strata of the subsoil, with the threefold advantage of being automatic, elegantly solving the problem of heat dissipation and minimizing the risks of soil contamination.

Parametric study of self-burial

It is obvious that the use of a self-burial technique, like any other storage method, creates numerous problems. To name only a few, the containers must be made of materials suited to the nature of the subsoil chosen for burial, care needs to be taken in filling and transporting the containers, which are sources of considerable radioactivity and heat, and sites have to be found in places where the subsoil is uniform.

These problems, although important, are not tackled in this preliminary study. The need to investigate them will depend on the answer to the question as to whether self-burial can in fact work using sources in the form of radioactive waste (limited thermal power and decay with time) and containers of feasible dimensions.

Accordingly we shall examine here only the influence of the parameters directly governing the self-burial phenomena, i.e.:

— the external configuration of the containers and their overall dimensions;

Table I: *Estimates of production and activity of radioactive waste in solid form in the Community.*

(a), (b): These figures are based on a minimum nuclear energy development programme (40 000 MWe in 1980, 400 000 in 2000) (1).

(c) : Volumes of waste accumulated at the end of each decade (1).

(d) : To give a tangible idea of the problems posed by storage, we have characterized waste by the heat it generates. The thermal power existing by the end of the decade is due to all the solid waste accumulated at that date. It has been calculated from American data (2) on the assumption that the waste is stored for one year after discharge and that reprocessing causes dilution of the pure fission products by a factor of 10.

	1970	1980	1990	2000
(a) Curies discharged per year ($\times 10^9$)	0.6	6	28	60
(b) Annual production (m^3)	13	130	500	1 200
(c) Cumulated volumes (m^3)	20	650	4 000	13 000
(d) Estimate of thermal power present (MW)	—	60	250	700

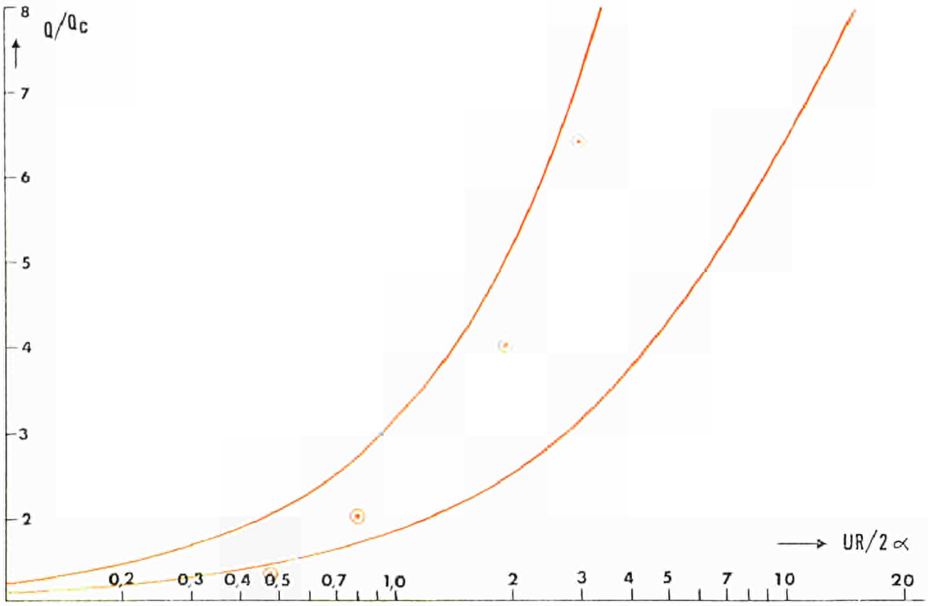


Fig. 1: *Burial rates U attainable with spherical containers are a function of: (a) radius R; (b) ratio between power Q generated by waste and critical power defined by equation [1]; (c) thermal diffusivity ϰ of soil chosen for burial. At equal available power, the burial rate of a container depends on the mode of distribution of the heat flux over the outer surface of the container. The right-hand curve in the figure is for an isothermal container, and the left-hand curve for uniform distribution of the heat flux over the outer surface of the container.* ○ *Experimental data on the burial of a steel sphere in paraffin.*

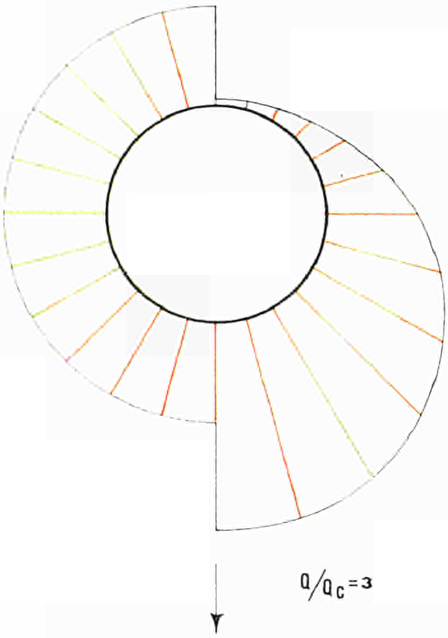
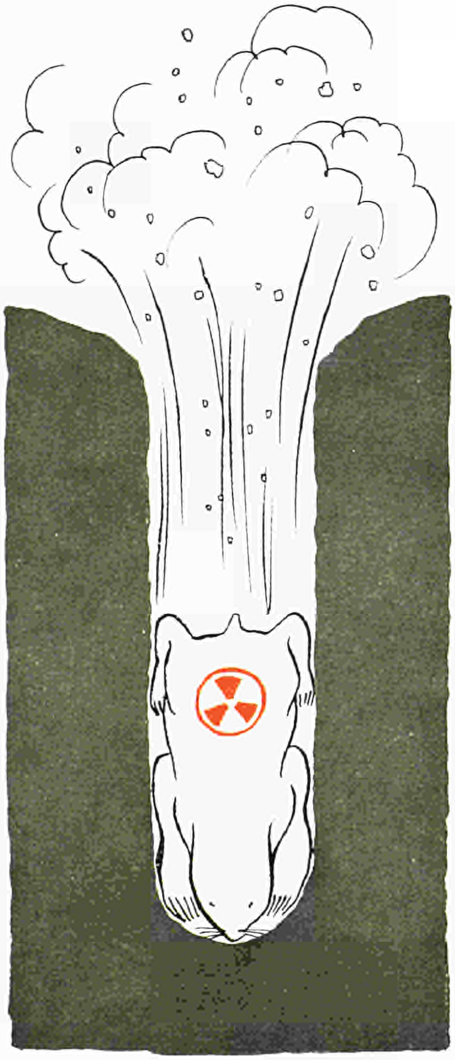


Fig. 2: *Isothermal sphere and uniform flux model. Illustration of the distribution of surface heat flux. The isothermal sphere model has a heat flux concentration in the burial direction and thus, at equal available power, gives a higher burial rate than the model with uniform flux distribution. (The arrow points at the direction of burial).*



- — the specific power of the heat source and its decay law;
- — the thermal properties of the subsoil.

Any solution to the self-burial problem involves an examination of the movement in the subsoil strata of a heat source of finite dimensions and time-dependent power output.

It will readily be realized that simplifying hypotheses are necessary to make the solution of such a problem feasible.

In order to linearize the differential equations governing the process, it is assumed that the thermal properties of the soil are independent of temperature.

It is also assumed that the temperature field round the container appears stationary to an observer at the heat source. This hypothesis entails a constant rate of burial in an infinite and homogeneous medium. As the power of the source decreases in time, the time must be broken up so that the power of the source can be considered as constant within each interval.

Finally, dipole sources due to melting and freezing of the soil and the effects of viscosity will be disregarded.

In a first analysis, the presence of the container was simulated by applying to the edges of a cylindrical or spherical cavity made in the ground boundary conditions either of uniform thermal flux

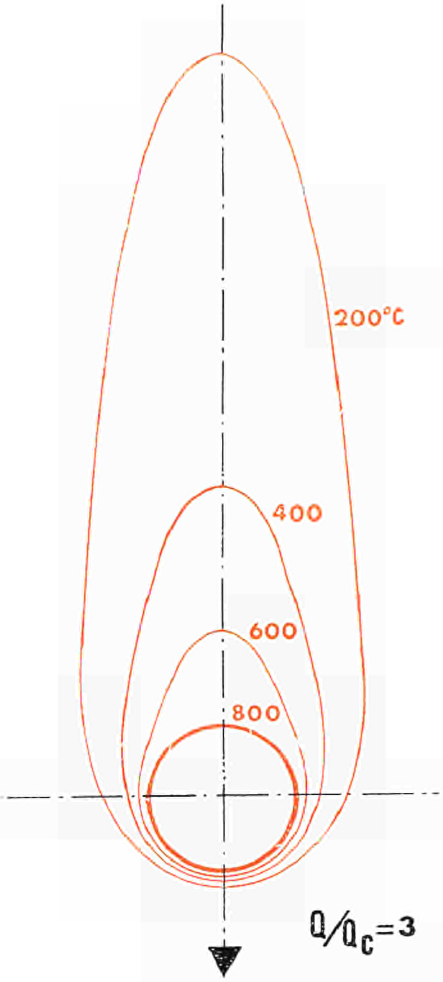


Fig. 3: *Distribution of isotherms round a sphere undergoing burial in salt, on the assumption that the sphere surface is at a uniform temperature equal to the melting point of the salt. (The arrow points at the direction of burial).*

or of uniform temperature equal to the melting point of the soil. This twofold analysis enables the top and bottom limits to the actual behaviour of a container to be defined and should permit a definition of the most interesting areas for a subsequent more detailed study.

Spherical containers

Critical powers — It can readily be demonstrated (4) that the minimum thermal power Q_{cr} which a spherical container of radius R (cm) must generate to initiate the self-burial process is given by the equation

$$Q_{cr} = 4 \pi R k T_m \text{ (watts)} \qquad [1]$$

where k (W/cm °C) is the thermal conductivity of the soil and T_m (°C) its melting point.

The self-burial of a spherical container holding radioactive waste of power density q (W/cm^3) is therefore conceivable only if

$$q > \frac{3 k T_m}{R^2} \qquad [2]$$

To establish an order of magnitude for the power densities involved, let us take the case of burial in salt, for which

$$k \simeq 0.06 \text{ W/cm °C}; \; T_m \simeq 800 \text{ °C}.$$

The minimum power densities are shown in Table II as a function of the container radius.

It is not very easy to determine the values of the power densities available in radioactive waste. These depend very much on the type of fuel, the burnup, the time elapsing between reactor shutdown and storage of the waste and also on the dilution of pure fission products achieved when the fuel is reprocessed.

The power densities can be estimated on the basis of a standard fuel described in an American study (2) and normally chosen as a reference for future reprocessing installations (3).

The reference reactor is a *PWR* burning UO_2 enriched with 3.3% U^{235}. It operates at a mean power level of 34.8 MW/t and has a burnup of 33 000 MWd/t.

The information in the work cited in the bibliography enables the specific power of the pure fission products to be evaluated as a function of the time elapsing since reactor shutdown. Thus, after 90 days of cooling, the pure products have a power density of 6 W/cm^3; after 150 days, the power density drops to 4.4 W/cm^3; after one year, it would be 2 W/cm^3.

Assuming that reprocessing dilutes the pure products by a factor of 10, radioactive waste of an initial power density equal to several times the final critical densities given in Table II should thus be available.

Burial rates — The burial rates obtainable with spherical containers are a function of their dimensions, the power density available and the thermal properties of the soil.

In fact, the problem is a fairly simple one, since all these parameters can be incorporated in two non-dimensional quantities, namely:

— the ratio Q/Q_{cr} of actual thermal power to the critical power defined by equation [1]:

$$Q/Q_{cr} = R^2 \cdot q/3 \, k \cdot T_m \qquad [3]$$

— the ratio:

$$Z = \frac{U \cdot R}{2 \alpha} \qquad [4]$$

of the product of the burial rate and the container radius divided by twice the thermal diffusivity of the soil.

The full investigation of self-burial of spherical containers thus reduces to the construction of a diagram giving the ratio Q/Q_{cr} as a function of the quantity Z.

It is, however, obvious that the value of Z (and therefore the rate) correspond-

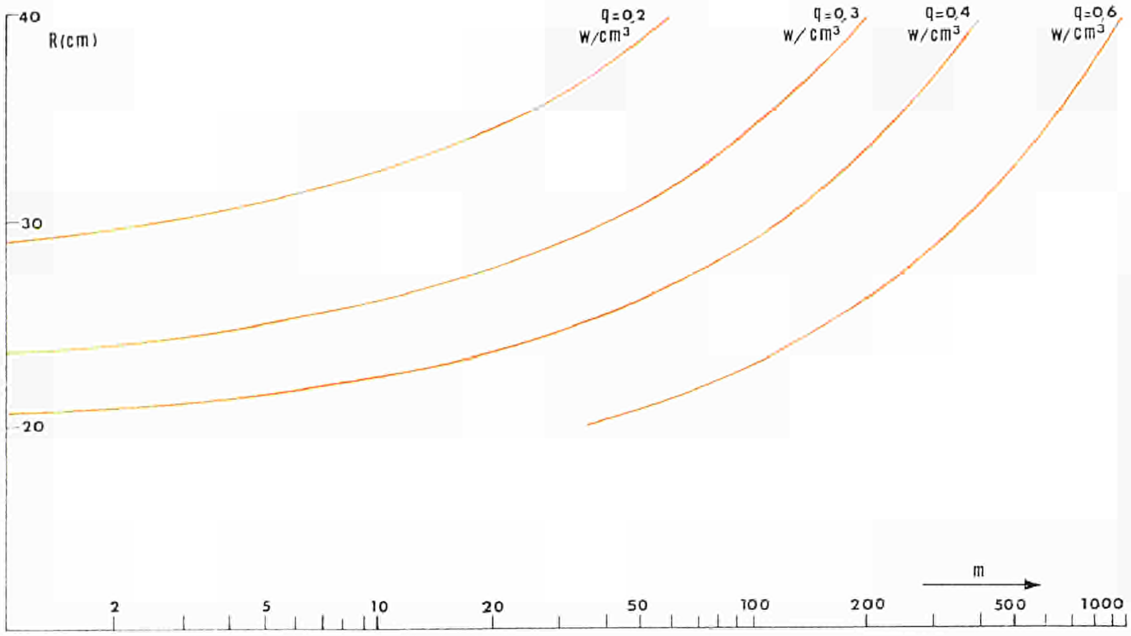


Fig. 4: *The curves show the self-burial depth attainable in salt. They are plotted as a function of the radius R of the spherical container and of the* initial *power density q of the waste. The sphere is assumed to be isothermal; the maximum depth is reached when the power density of the waste is reduced to a value equal to the critical density defined by equation* [2].

ing to a given value of the ratio Q/Q_{cr} depends on the way in which the heat flux is distributed over the surface of the container in contact with the soil. As was previously mentioned, two configurations have been devised in order to describe the actual behaviour of a spherical container.

In the first configuration, it is assumed that the edge of the spherical cavity simulating the container is at a uniform temperature equivalent to the soil melting point T_m, whereas at a considerable distance away the temperature remains undisturbed.

The numerical resolution of the temperature field for various values of the ratio Z, followed by integration of heat flux to obtain the power generated by the container, gave the behaviour described by the right-hand curve in Fig. 1.

The second configuration assumes a heat flux uniformly distributed round the edge of the spherical cavity. The results obtained lead to the behaviour law described by the left-hand curve in Fig. 1.

It will be observed that, for equal power, the burial rate of an isothermal sphere is much higher than that of a sphere with uniform flux and the same diameter. The explanation is that, unlike the uniform flux model, the isothermal sphere model has a heat flux concentration in the direction of burial. Fig. 2 shows the heat flux distribution for the two models in question, while Fig. 3 shows the distribution of isotherms round a sphere in motion, it being assumed that the sphere surface is at uniform temperature.

Burial depths — The diagrams in Fig. 1, expressed in polynomials, contain all the information required for calculating the burial depths obtainable as a function of container radius and initial power density.

In this calculation, account was taken of the decay in time of the thermal power from the waste, by a process of numerical integration at short time intervals during which the burial rate may be regarded as constant.

The burial depths obtainable were calculated on the assumption that the subsoil consists of salt.

Salt was not chosen at random. Firstly, geologists tell us that there are numerous salt domes in Europe several miles in diameter and more than 1 000 m high (7). Secondly, the melting point of salt is 800 °C, whereas that of other rocks, such as granite, is often over 1 500 °C. It should also be noted that salt domes are integral geological structures, i.e., they contain no faults or other defects. This characteristic is due to the fact that salt is viscous under pressure.

Table II: *Critical power densities as a function of radius of spherical containers. Burial in salt is assumed (melting point 800 °C). Self-burial is possible only if the power density of the waste exceeds the values given. It should be noted that, for containers with a radius of over 30 cm, the critical power densities are very low.*

R (cm)	10	20	30	40	50
q_{cr} (W/cm^3)	1.44	0.36	0.16	0.09	0.06

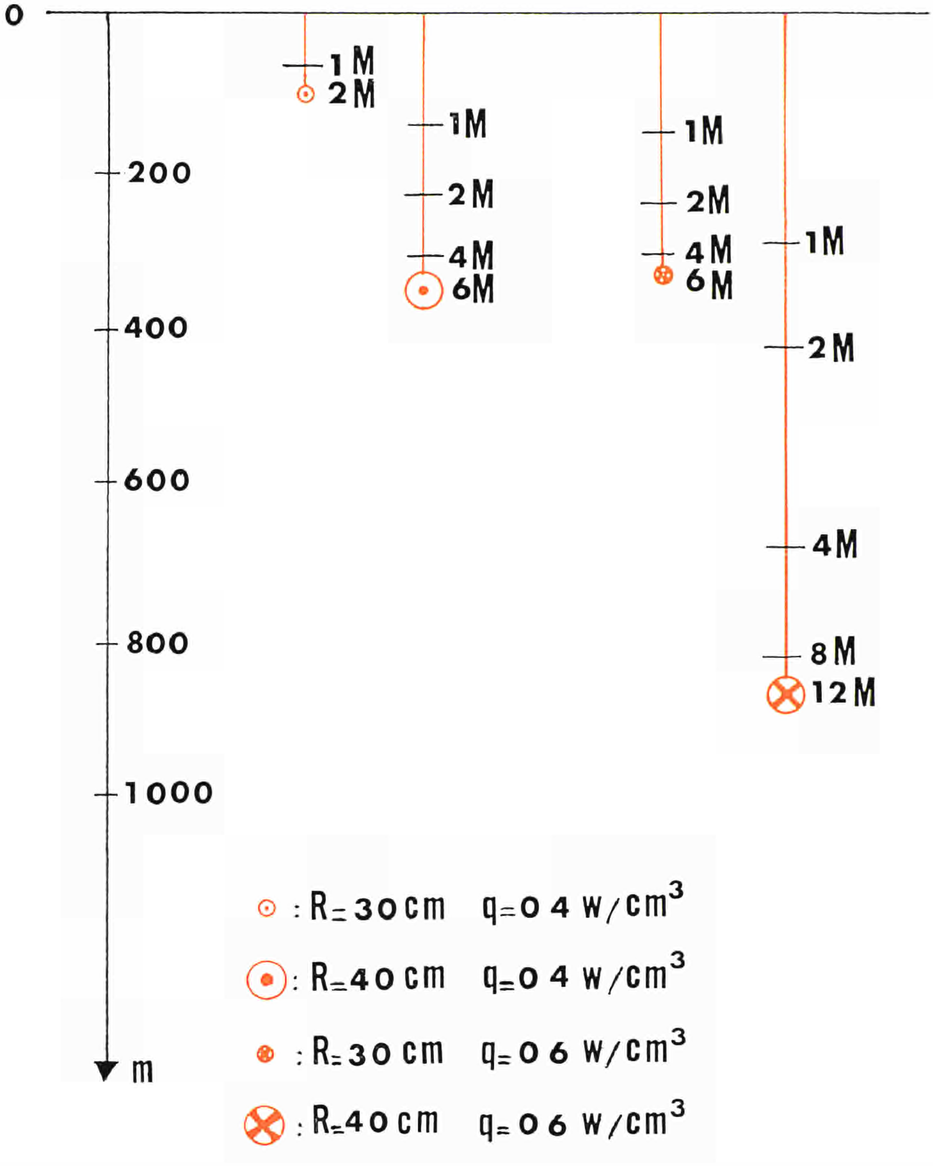


Fig. 5: *The dynamics of the self-burial phenomenon are illustrated by the time curves for the depth reached by four spherical containers of which the radii R and the initial power densities q are given [M month(s)]. The isothermal sphere model is used; burial takes place in salt.*

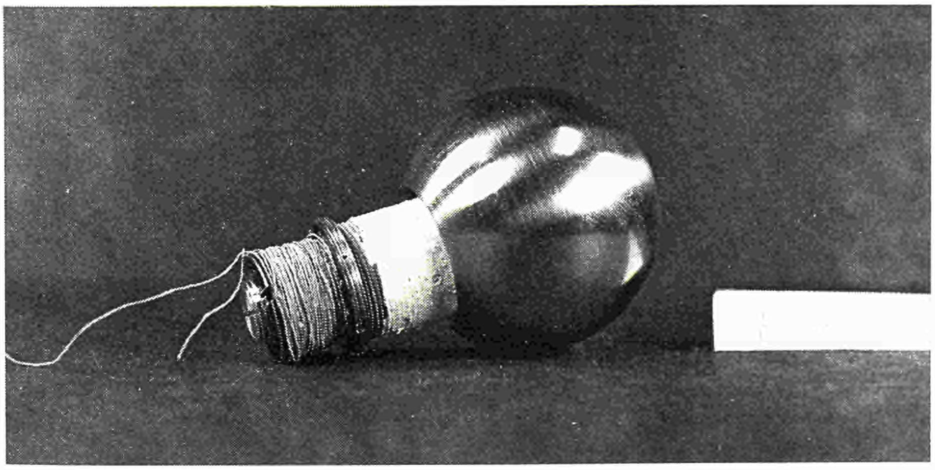


Fig. 6: *A preliminary check on the theoretical results was carried out by burying a steel sphere 5 cm in diameter in a paraffin block. The thermal power was supplied by the Joule effect of a resistor at the centre of the sphere with an external power supply. The results obtained are shown in Fig. 1; they are in fairly good agreement with the estimates of the isoflux model, as might have been expected from the type of experimental device used (low level of internal conduction).*

Fig. 4 shows the self-burial depths obtainable in a salt dome. In order to visualize the dynamics of the phenomenon, Fig. 5 shows the penetration in depth as a function of time. The diagrams refer to isothermal spheres of a radius of 30-40 cm; the initial power density of the waste is 0.4 or 0.6 W/cm^3.

Experimental verification — In order to check the theoretical data on the burial rates attainable, we carried out an experiment involving the self-burial of a stainless steel sphere 5 cm in diameter in a block of pressed paraffin. The thermal power required is obtained, as Fig. 6 shows, by the Joule effect in an electrical resistor at the centre of the sphere. This resistor is supplied by an external source, the wiring being passed through a zone of liquid paraffin above the sphere.

After an experimental determination of the critical power, we measured the burial rates for various values of the ratio Q/Q_{cr}. The experimental points are plotted in Fig. 1; it will be noted that they lie well inside the area delineated by the theoretical curves.

Cylindrical containers

There are three main phases in the study of the burial of cylindrical containers, as in the case of spherical containers:

- calculation of critical powers;
- evaluation of burial rates as a function of power available, geometry and subsoil characteristics;
- estimation of burial depths attainable.

Only the case of a vertical cylinder has been considered, i.e., with its axis of rotation parallel to the direction of burial.

It is shown (6) that the minimum thermal power which has to be generated by a cylindrical container of radius R (cm) and height/diameter ratio equal to l to start the self-burial process can be found from the equation

$$Q^{c}_{cr} = 4 \pi R k T_m \cdot$$

$$\cdot \frac{2l}{\log_e [2l + \sqrt{1 + 4l^2}]} \text{ Watts} \quad [5]$$

Compared with equation [1], this expression shows that the critical power of the cylinder is equal to that of a sphere of equivalent diameter multiplied by a form factor greater than unity and increasing with the value of the height/diameter ratio (see Table III).

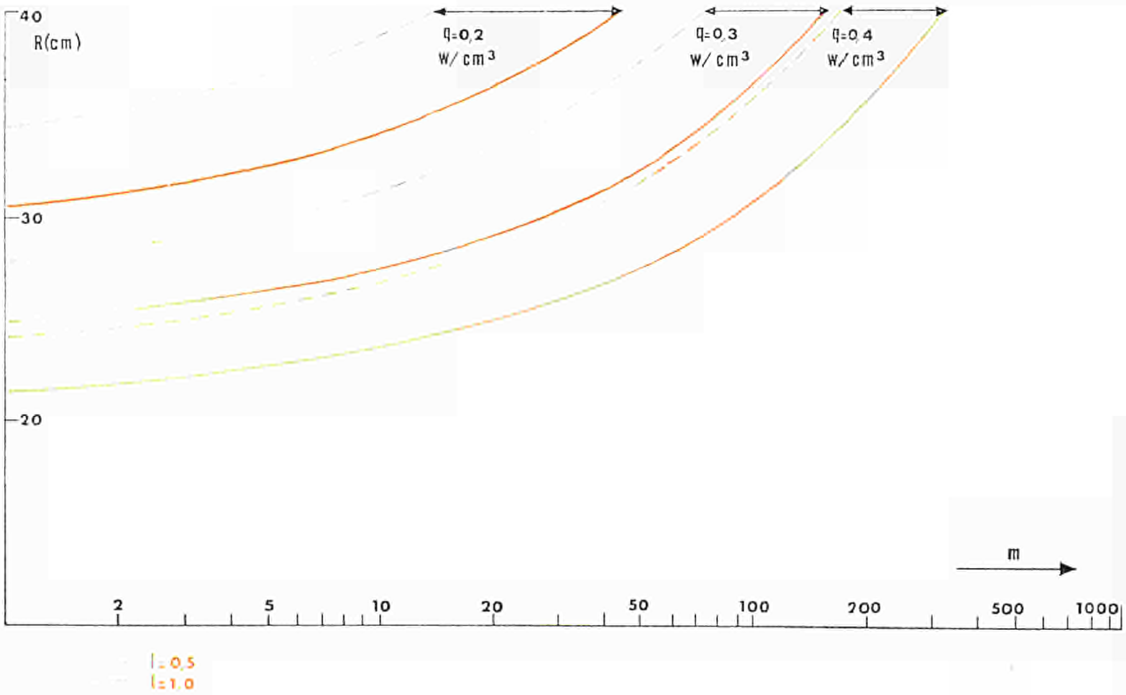
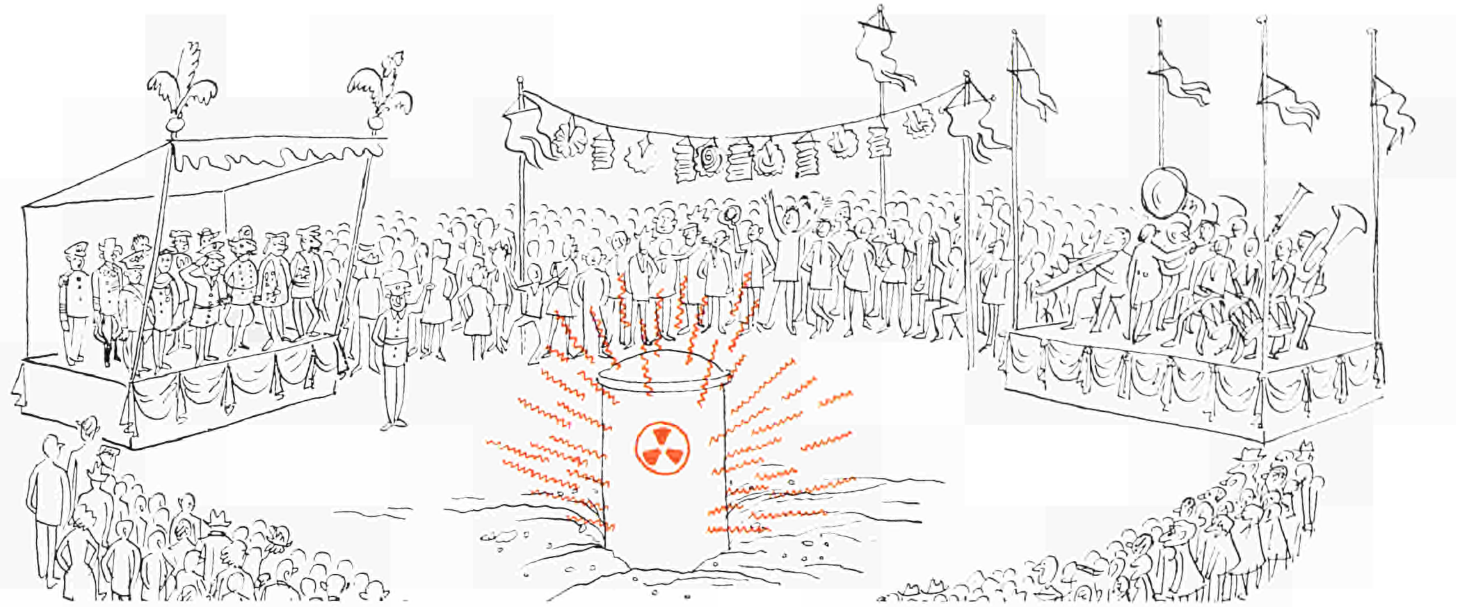


Fig. 7: *Burial depths attainable in salt with cylindrical containers. Two values (0.5 and 1) of the height/diameter ratio were considered. The cylinder surface is assumed to be isothermal; the* initial *power density of the waste is shown.*



The burial rates are obtained by plotting diagrams giving the ratio Q/Q^{c}_{cr} (actual power/critical power) as a function of the parameter Z defined in equation [4] and the ratio l. Only the isothermal model was used in order to approximate the behaviour of a real cylinder.

The burial depths obtainable were calculated for salt and for height/diameter ratios of 0.5 and 1. Fig. 7 shows the depths obtained for the two cylinder types as a function of radius and of the initial power density of the radioactive waste.

Conclusions

A preliminary theoretical study, confirmed by laboratory experiments, has shown that the self-burial phenomenon is a fact.

Table III: *Form factor affecting the critical power of a cylindrical container.*

	Form factor
1	1.39
2	1.90
3	2.41
4	2.88

The power densities involved are of an order which allows the use of such a process to be contemplated for the definitive storage of high-activity radioactive waste.

The thorny problem of the removal of the heat generated by radioactive decay is elegantly solved by using this heat as a driving force for the self-burial process tiself. It even becomes desirable for the fission products accumulated in solid form not to be cooled down or diluted too much, so that significant burial depths can be achieved with capsules of acceptable dimensions.

At the same time, it is obvious that we cannot claim, on the basis of the arguments put forward in this article, to have solved all the problems connected with the application of a means of disposing of high-activity waste by self-burial.

What we have tried to do rather is to point out a promising line of research.

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References: (1) G. GRISON: The creation of a Community system of radioactive waste dumps. *euro-spectra*, 10 (1971) No. 3, pp. 91-94. (2) "Siting of fuel reprocessing plants and waste management facilities", Report compiled and edited by the Staff of the *Oak Ridge National Laboratory*, July 1970. (3) E. DUTILLEUX, Eurochemic, Mol, Belgium, personal information. (We acknowledge with gratitude Mr Dutilleux's interest in our work and the valuable information he has provided.) (4) C.R. EASTON: "Conduction from a finite-size moving heat source applied to radioisotope capsule self-burial", (Douglas Aircraft Company, Santa Monica, California) pp. 209-218 of Thermal Conductivity, Flynn *et al.*, editors, Washington, *National Bureau of Standards*, 1968. (The power for thermoelectric generators on board satellites usually obtained from radioisotope sources. The study in question was undertaken to estimate the consequences of fall-out from a capsule of radioisotopes in the event of an abortive launching). (5) G. de BENI, C. MARCHETTI: Hydrogen, key to the energy market. *euro-spectra*, 9 (1970) No. 2, pp. 46-50. C. MARCHETTI: Hydrogen, master-key to the energy market, *euro-spectra*, 10 (1971) No. 4, pp. 117-129. (These articles show how nuclear energy can cover not only electric power requirements but also most requirements of the energy market). (6) J. DONEA, S. GIULIANI, L. KESTEMONT: Process of self-burial of containers holding high-activity nuclear waste *Report*, *EUR 4903 f* (1972). (7) H. KRAUSE *et al.*: Project for storing radioactive wastes in a salt cavity. Proceedings of the Symposium organized by the *IAEA* and *ENEA* on the Disposal of Radioactive Wastes into the Ground, Vienna, 29 May-2 June 1967. Published by the *International Atomic Energy Agency*.