UCRL - 73130
PREPRINT
Conf-711105--1

Lawrence Radiation Laboratory
UNIVERSITY OF CALIFORNIA
LIVERMORE
94550

NUCLEAR CHIMNEYS: UNDERGROUND CONTAINERS FOR CHEMICAL AND METALLURGICAL PROCESSING

A. E. Lewis

August 26, 1971

This paper was prepared for presentation at the Symposium on the Underground Environment sponsored by the Geological Society of America, Washington, D.C., November 3, 1971

Nuclear Chimneys: Underground Containers for Chemical and Metallurgical Processing*

Arthur E. Lewis

Lawrence Livermore Laboratory, University of California
Livermore, California 94550

ABSTRACT

It is practical to use contained nuclear explosions to construct underground openings (nuclear chimneys) under a variety of geologic conditions. The underground environment of these chimneys provides opportunities for the development of industrial processes that may not be practical at the earth's surface. Examples to be discussed here are: the *in situ*, high-pressure, high-temperature recovery of metals from sulfide ores, and the *in situ* incorporation of nuclear-fuel reprocessing waste in molten silicate rock.

INTRODUCTION

It has proved to be more difficult and expensive to work in an underground environment than at the surface. Man has been forced underground by his need to obtain raw materials and energy. We obtain

---

*Work performed under the auspices of the U.S. Atomic Energy Commission.

oil, gas, and geothermal steam by drilling into natural reservoirs. We obtain natural brine solutions, sulfur, and soluble minerals from other drill holes. Ores are obtained from mines and brought to the surface so that their metal values may be extracted. Waste material, either solid or liquid, is often put back into the mines or into permeable formations.

In all these examples material is extracted from the earth or deposited in the earth. Since processing is done almost entirely at the surface no advantage is obtained from the characteristics of the underground environment. In mining, the cost of moving the ore is a major limitation on the depth and grade of ore that may be mined.

The chemical and physical properties of rock, together with the temperatures and pressures (both hydrostatic and lithostatic) that prevail underground, offer advantages yet unused.

One difficulty in using these advantages is the expense of finding or constructing suitable space. The use of nuclear explosives which may be emplaced through drill holes and detonated safely and at relatively small cost compared to other methods, may provide suitable space for underground processing, as well as a means of fracturing ore bodies prior to further processing. Before several examples of such processes are given, the effects of deeply buried nuclear explosives will be reviewed briefly.

NUCLEAR EXPLOSIVE EFFECTS[1,2]

When a nuclear explosive is detonated underground, a large amount of energy is deposited in a short time by the nuclear reaction. The high temperatures and pressures produced are sufficient to vaporize the device and some of the surrounding rock. This vapor is the working gas which expands against the confining pressure of the overlying rocks to produce a more or less spherical cavity. The size of the cavity is a function of explosive energy released (yield), depth of burial, and rock properties.

The rock gas condenses during or soon after the expansion of the cavity. As the temperature and pressure fall, the roof of the cavity usually collapses. The collapse proceeds upward until the roof becomes self-supporting or until the space is filled with broken rock. The result is a column of broken rock called a chimney, with the volume of the cavity distributed through it as open space. The cavity-chimney formation history for a 5-kt explosion in granite is shown in Fig. 1. In a plastic material such as salt, a standing cavity which does not collapse to form a chimney may be produced.

The average temperature in a chimney of broken rock may be raised only a few degrees in a shallow detonation that produces a large cavity and a large chimney. At greater depth, the same explosive will deposit approximately the same amount of heat, but it will be distributed in a smaller chimney and the temperature will be higher, perhaps many hundreds of degrees centigrade hotter.

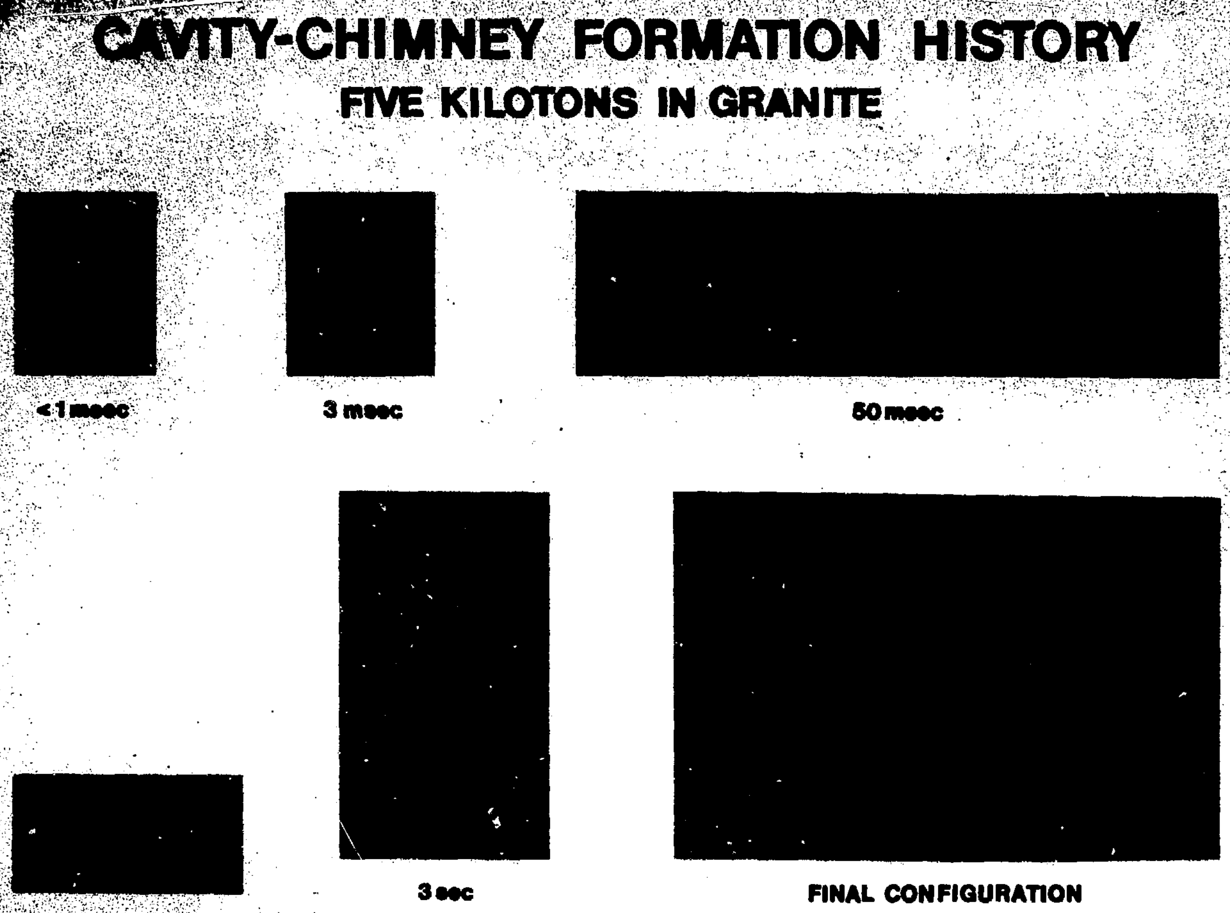


Fig. 1. Cavity-chimney formation history—5 kt in granite.

Therefore, the starting condition for any application of underground nuclear explosions is a cavity or a column of broken rock surrounded by a fracture region of varying extent (a few cavity radii) around the detonation point. The starting temperature in the chimney will vary from tens to hundreds of degrees centigrade above ambient, depending on the depth of the explosion and the properties of the rock.

If the chimney and fracture region are in low-permeability rock, the chimney may be used as a pressure vessel. If the permeability is high, the pressure will be limited to the formation pressure or the hydrostatic pressure if the chimney fills with water.

CHEMICAL MINING OF COPPER[3,4,5]

Copper is one of the metals whose ores occur in deposits of large enough size to permit economic breakage with nuclear explosives. The prospect of leaching the copper from the *in situ* broken ore would be of great interest if universally applicable. The primary copper mineral in most large copper deposits is chalcopyrite, a mineral that is only very slowly dissolved by the sulfuric acid or ferric sulfate systems presently used for leaching. Leaching of copper ores is therefore generally limited to ores that have been oxidized and/or enriched by natural processes. This process consists of the oxidation of iron and copper sulfides by the oxygen contained in water percolating downward from the surface. The copper may be deposited as oxides, sulfates, carbonates, and silicates above the water table or carried downward, where it is reprecipitated under more reducing conditions as a secondary sulfide, usually chalcocite or covellite.

These deposits of oxidized copper sulfides and soluble secondary sulfides generally occur near the surface, say within 1000 ft. While many of these may be broken by conventional or nuclear explosives and leached *in situ*, future copper supplies must come more and more from the deep primary deposits. At present, removal by conventional mining methods and metallurgical processing at the surface provide the only means available for these primary sulfide ores. The costs increase with depth and decreasing grade of ore.

Properties of the underground environment, such as high hydrostatic pressure and good thermal insulation, can be utilized in a process that we believe will make *in situ* chemical mining of deep chalcopyrite deposits practical.

A contained underground nuclear explosion is used to produce a chimney of broken ore below the water table (Fig. 2). The assumptions for this example are shown in Table 1. This chimney is allowed to fill with water until it is in hydrostatic equilibrium with the water table. Oxygen is then introduced into the bottom of the chimney at a pressure slightly higher than the hydrostatic head in the chimney. Because the solubility of oxygen in water is a function of its partial pressure, the amount of oxygen that may be dissolved is greatly increased, compared to the amount dissolved in equilibrium with air at 1 atm. The effect of increased oxygen solubility on the dissolution rate of chalcopyrite is

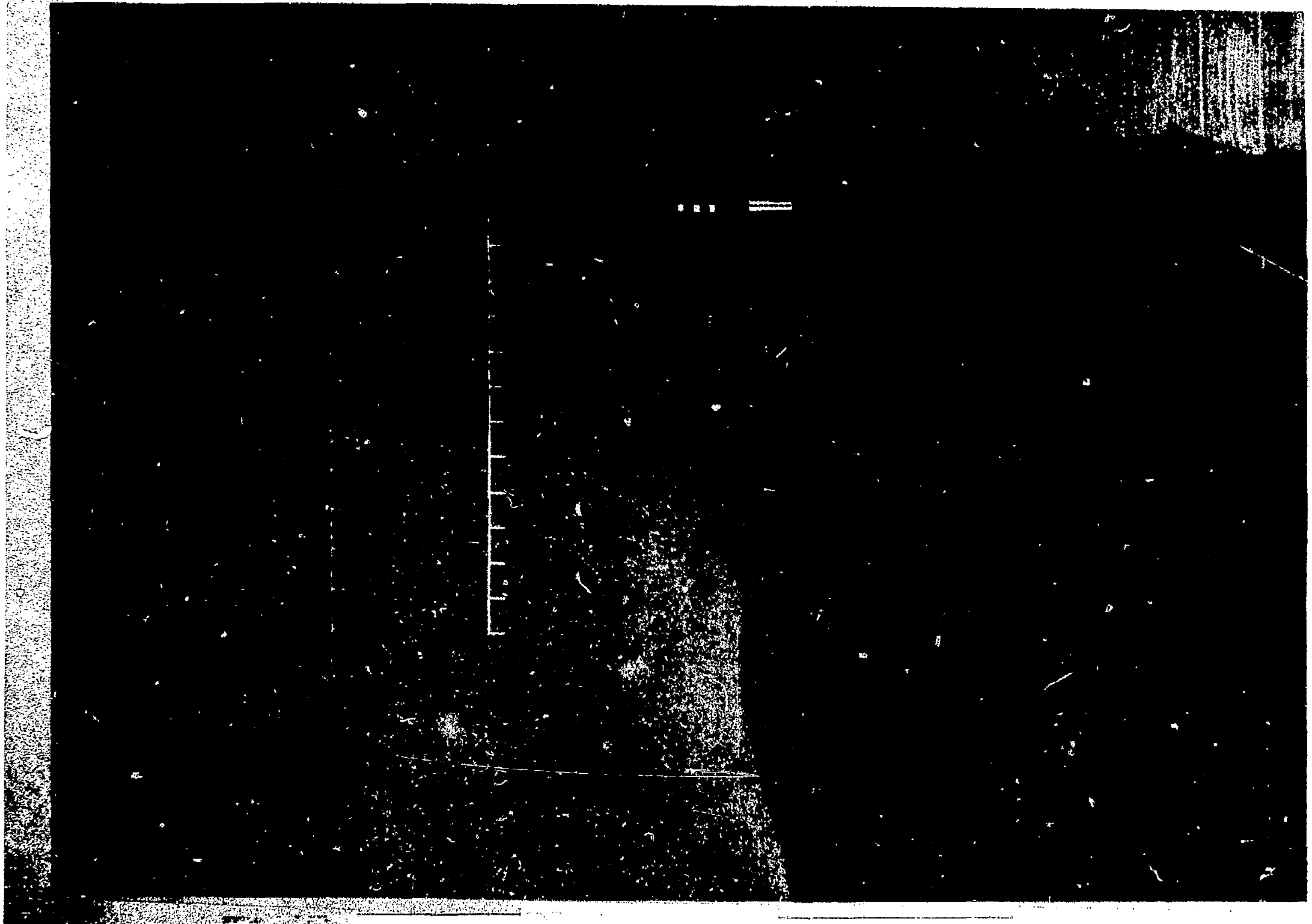


Fig. 2. Chemical mining—flooded primary sulfide ore.

Table 1. Chimney properties.

Yield of nuclear device	100 kt
Depth of burial	750 m
Depth of water table	50 m
Water content of rock	3 wt%
In situ density of rock	2.7 g/cm^3
Bulking porosity	0.17
Radius of cavity	41 m
Height of chimney	300 m
Volume of water in chimney	2.9×10^5 m^3
Volume of rock in chimney	1.4×10^6 m^3
Mass of rock in chimney	3.8×10^6 metric tons
Hydrostatic pressure at depth of burial	1000 psi
Hydrostatic pressure at top of chimney	570 psi

illustrated in Fig. 3. The increase in rate of dissolution of chalcopyrite as the oxygen pressure is increased is apparent at this temperature. By measurement of the surface area of chalcopyrite and a comparison of these values with experimental results, it can be shown that the rate of dissolution is a function of surface area. Rates of chalcopyrite dissolution per unit surface as a function of temperature are shown in Fig. 4.

The products of the decomposition of chalcopyrite are cupric sulfate, sulfuric acid, sulfur, and hematite. Pyrite in the system produces basic

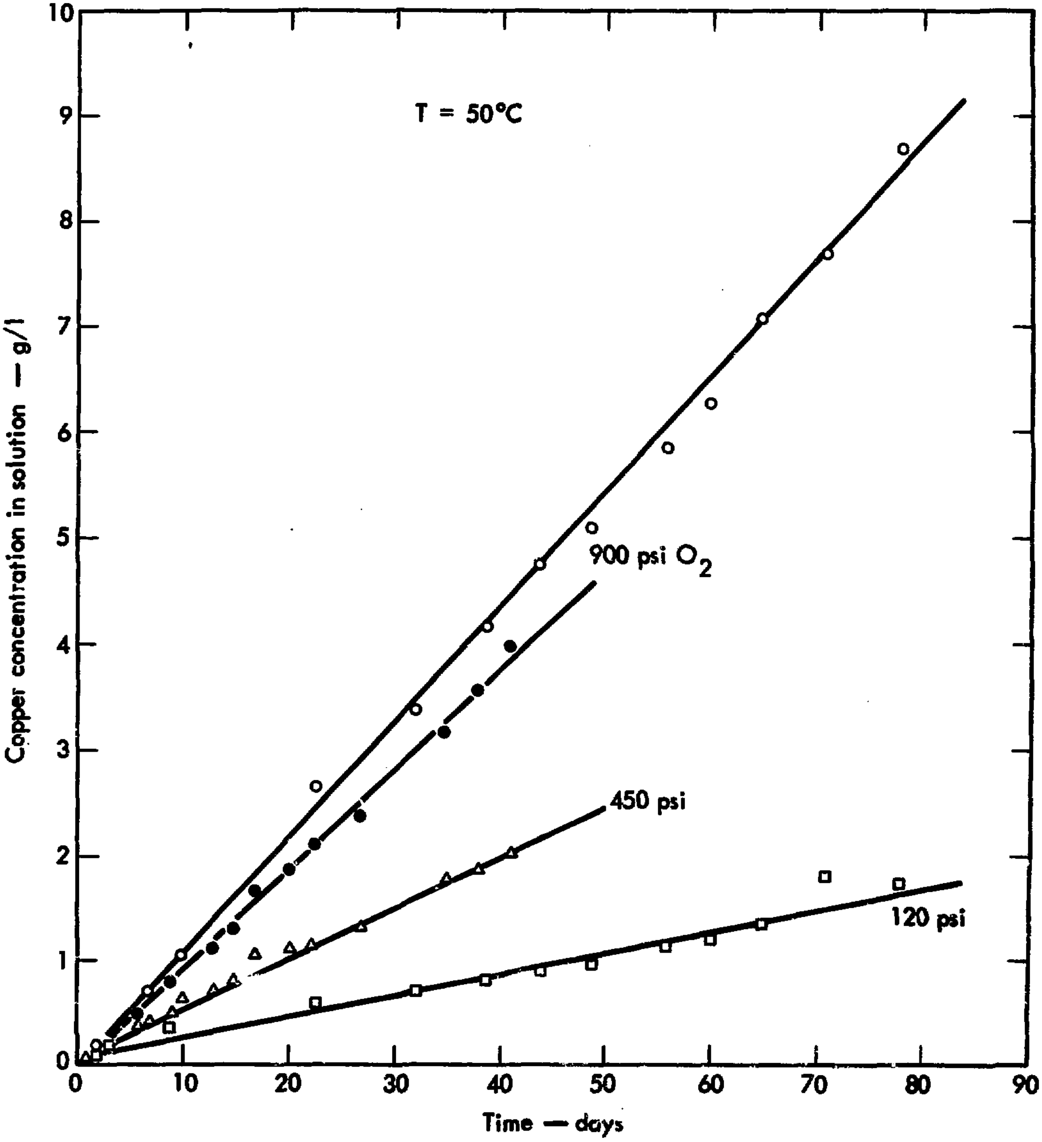


Fig. 3. Copper concentration in solution vs time.

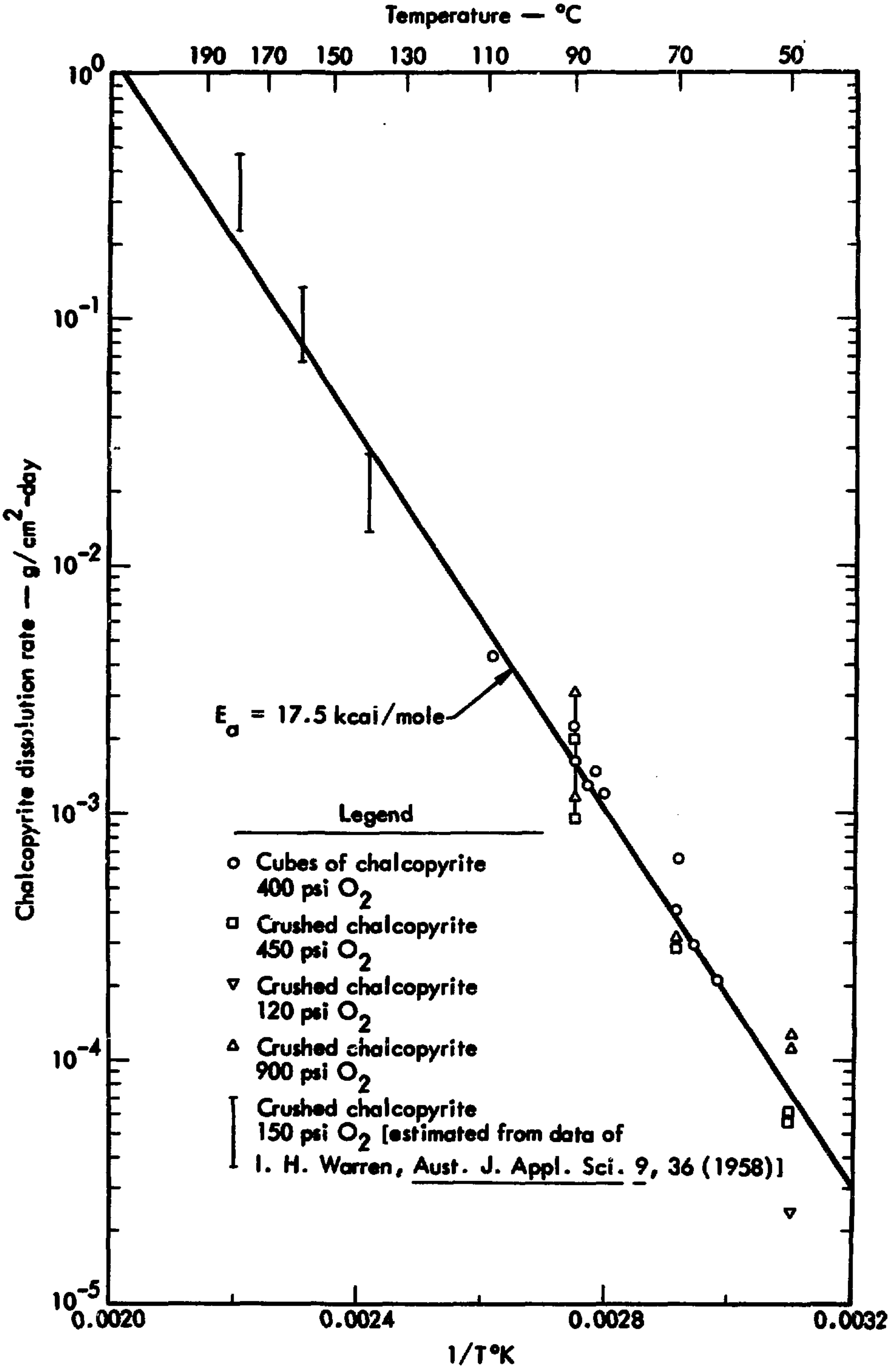


Fig. 4. Dissolution rate of chalcopyrite in dilute H_2SO_4 solution containing dissolved O_2.

iron sulfate, sulfur, and sulfuric acid. The pH soon stabilizes in the range of 1.5 to 2.5 with no net sulfuric acid production, and iron is almost absent in the solution.

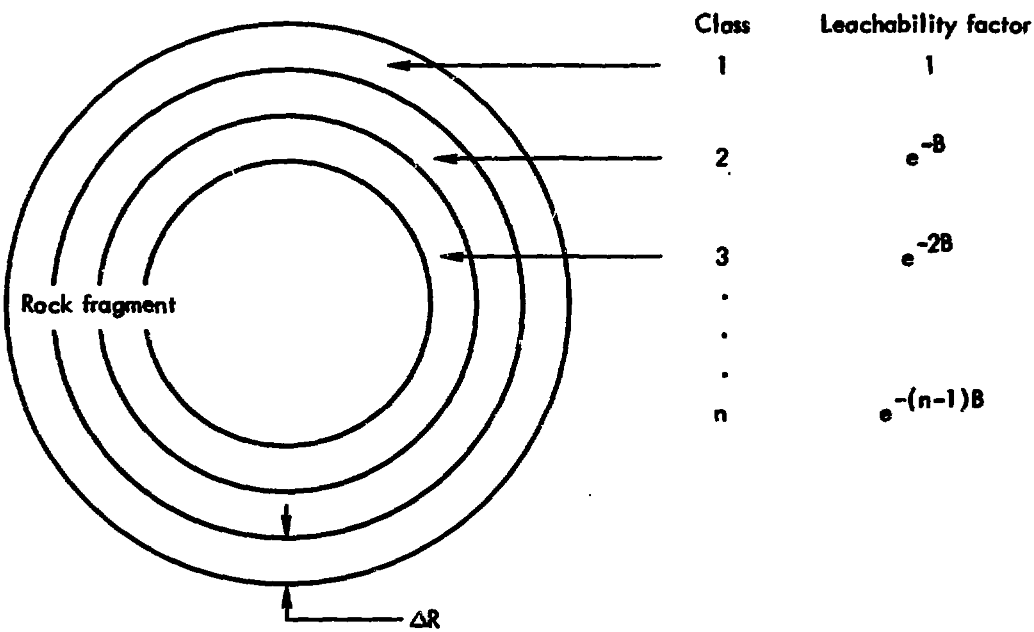
All these reactions are highly exothermic and will produce high temperatures at the underground conditions expected. The net result is that the rate of recovery of copper is not limited by the rate of dissolution of chalcopyrite, but by the rate at which copper may be removed from the interior of large blocks of ore.

A model for recovery from large rock fragments is illustrated in Fig. 5. The model is consistent with the removal of copper from the fragment by diffusion processes. The constants B and ΔR are being determined experimentally by measurement of the rate of recovery from various-size rock fragments. Results from small fragments (<2-cm diam) leached in the laboratory, and preliminary results from the leaching of larger fragments (<30-cm diam) in a 1000-gal pressure vessel indicate that the following constants are reasonable for the porphyry copper ore being studied:

$B = 0.7$, $\Delta R = 0.3$ cm, and chalcopyrite particle diameter = 0.2 cm.

A computer code has been developed which calculates the temperature in the chimney, the concentration of copper in solution, and the fraction of copper recovered as a function of time for the following variables:

- Initial temperature.
- Degree of oxygen saturation.
- Pyrite to chalcopyrite ratio.
- Grade of ore.



$$K'_{cp} = D \cdot K_{cp} \sum_{i=1}^{n} S_i e^{-(i-1)B}$$

where K'_{cp} = net rate of oxidation of chalcopyrite

K_{cp} = specific rate of oxidation of chalcopyrite

D = degree of saturation of solution with dissolved oxygen

S_i = total surface area of all chalcopyrite particles in class i

$e^{-(i-1)B}$ = leachability factor of chalcopyrite in class i

Fig. 5. Model for copper recovery from rock fragments.

The calculation takes into account such factors as:

- The rate of chalcopyrite dissolution as a function of temperature.
- The heat generated in the chimney by the exothermal reactions.
- The conductive heat loss from the chimney.
- The rate of removal of chalcopyrite from ore fragments.
- The particle-size distribution of ore fragments in the chimney.
- The dimensions of the chimney.

Figure 6 shows the chimney temperature as a function of time for three ore grades for an initial temperature of 60°C, a pyrite-chalcopyrite ratio of 1:1, and an oxygen saturation of 0.5. Figure 7 shows the fraction of copper recovered for the same set of conditions. It is apparent that it is essential to obtain a high temperature in the chimney in order to obtain high recovery of copper.

If the ore grade is 0.5 wt% copper (Fig. 8), the fraction of recovery increases as the amount of pyrite present increases. The increase results from the additional temperature rise produced by oxidation of pyrite. If the ore grade remains at 0.5 wt% copper and the pyrite-chalcopyrite ratio is 1:1, the fraction recovered may be increased by preheating the chimney. Figure 9 shows the effects of heating the chimney to initial temperatures of 50, 60 and 70°C. Such heating can be economically accomplished by injecting high-pressure steam.

This is one illustration of how nuclear technology and the underground environment itself can be used for metallurgical processing that would not be practical at the surface.

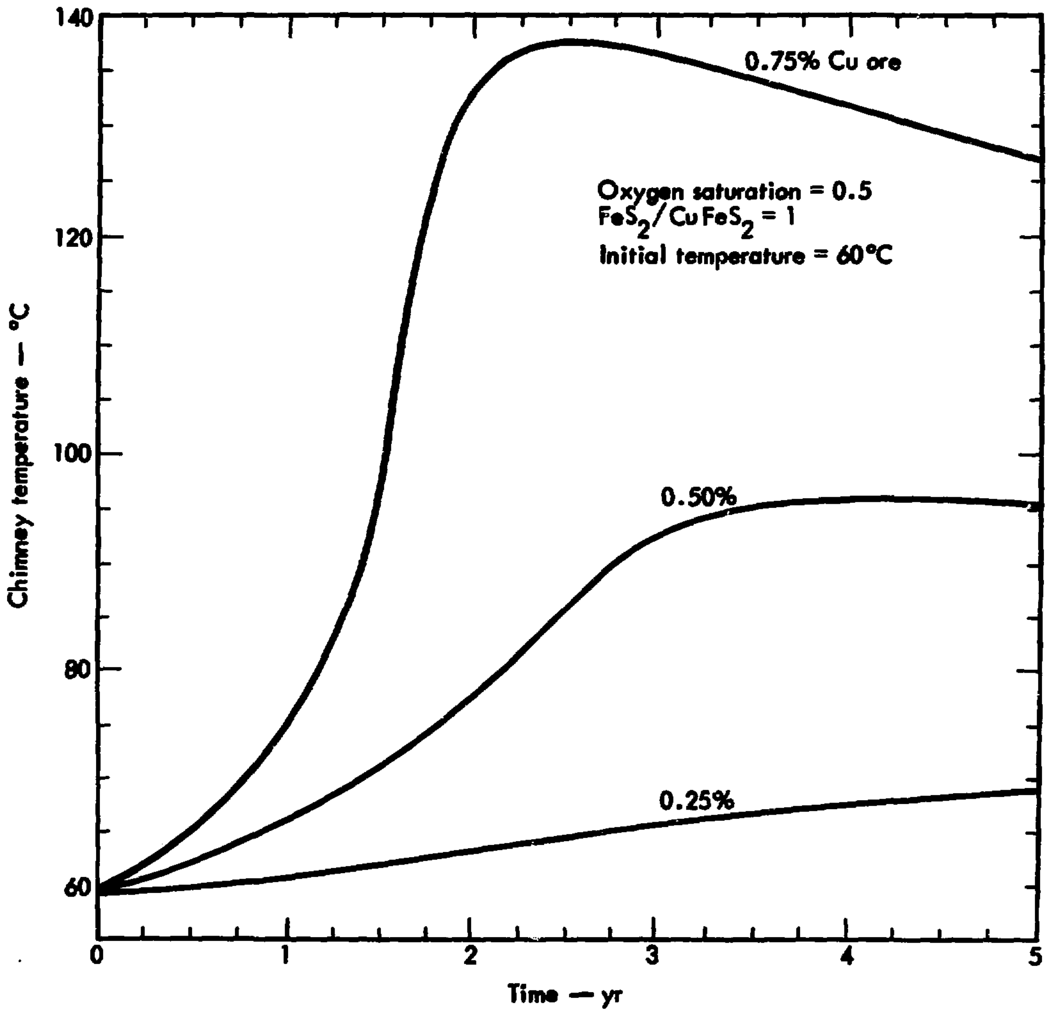


Fig. 6. Chimney temperature vs time for various grades of copper ore.

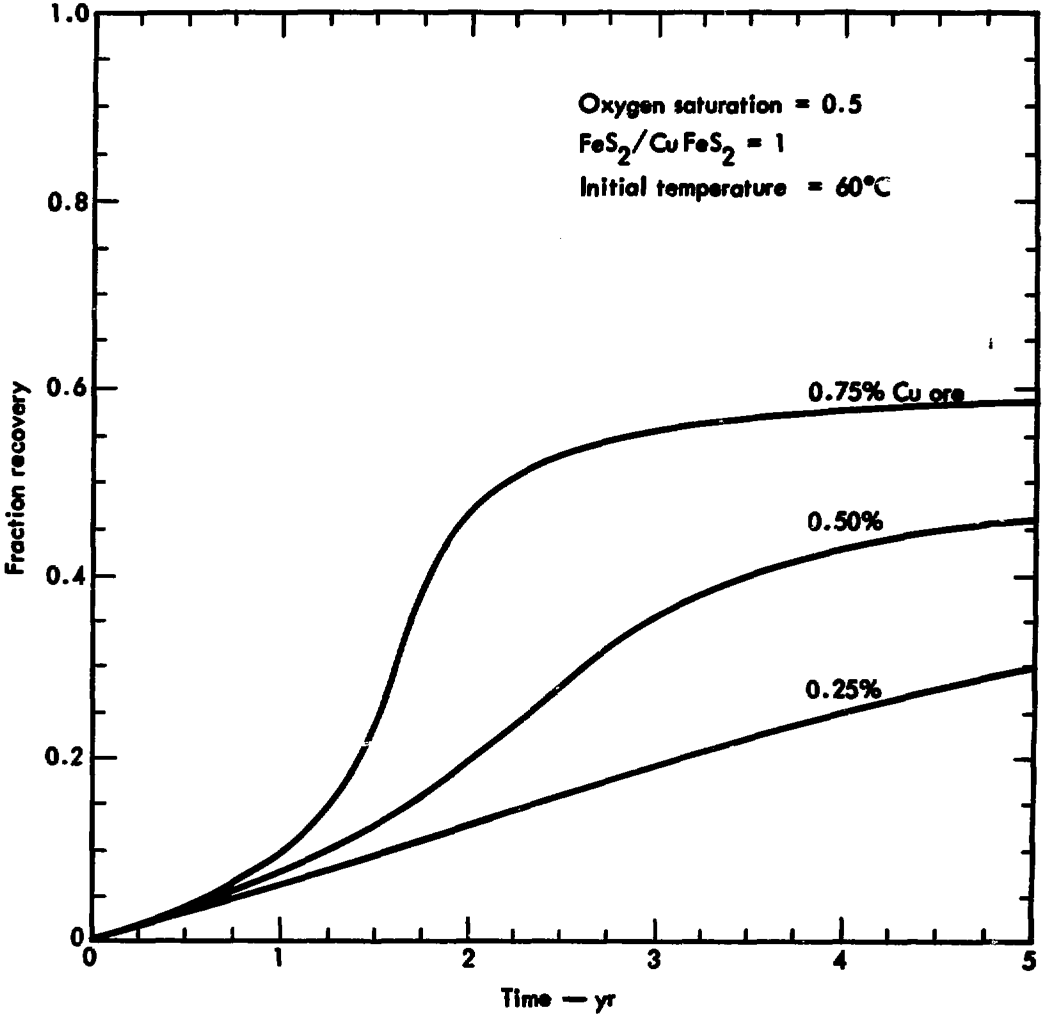


Fig. 7. Fraction recovery vs time for various grades of copper ore.

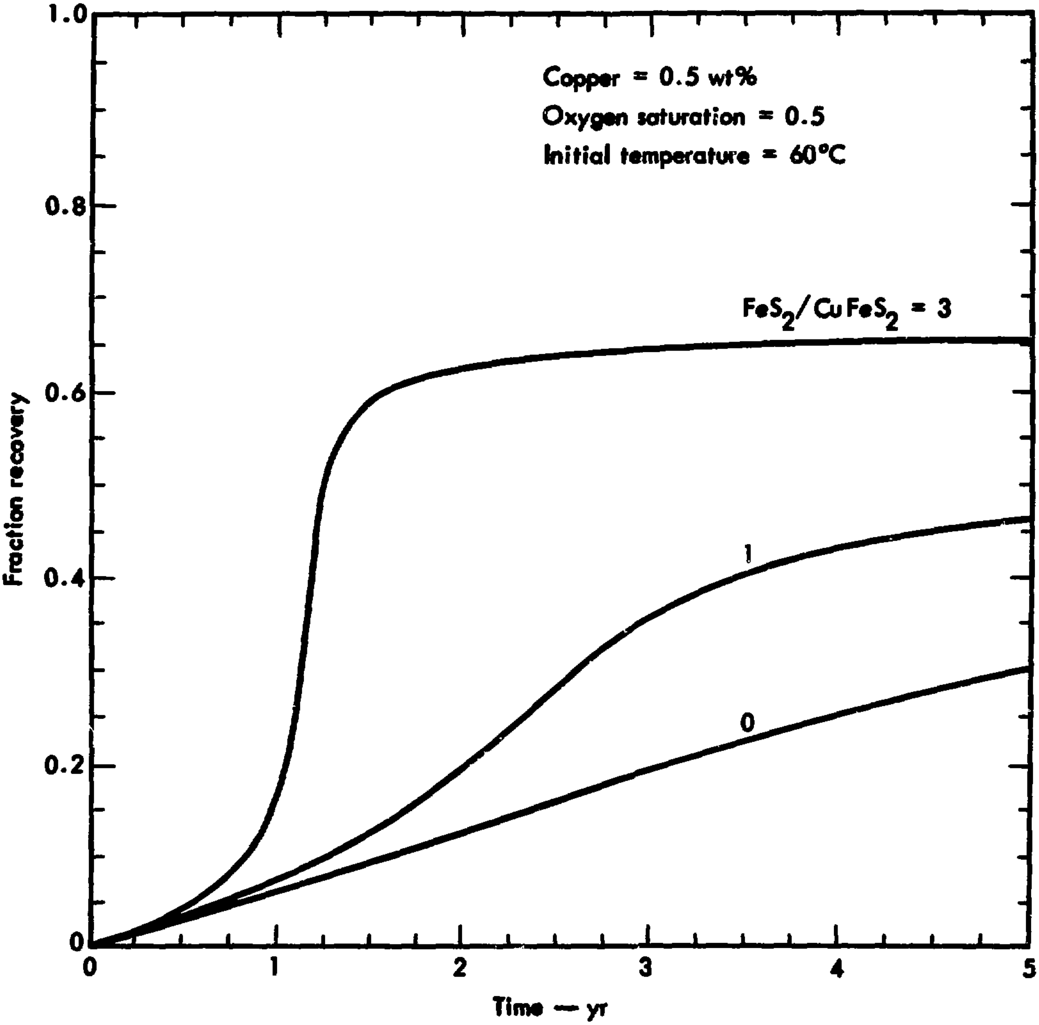


Fig. 8. Fraction recovery vs time for various $FeS_2/CuFeS_2$ mole ratios.

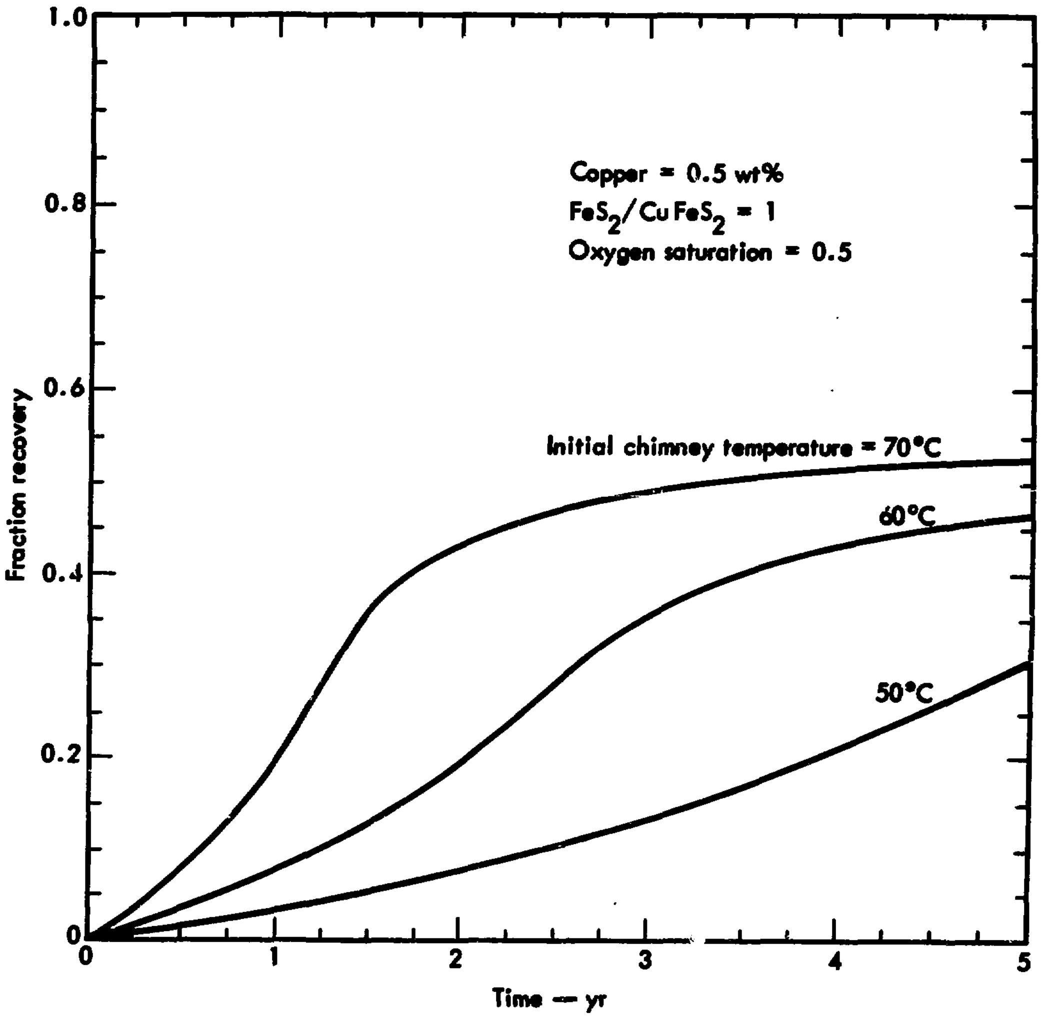


Fig. 9. Fraction recovery vs time for various initial chimney temperatures.

DISPOSAL OF WASTES FROM NUCLEAR REACTORS[6,7]

One of the problems that remains in the development of nuclear power is the management of radioactive wastes from nuclear-fuel reprocessing plants. The best solution[8] at the present state of the art appears to be to remove the wastes from solution, package them in a solid form, and transport them to a centralized repository for permanent underground emplacement. Because of their highly hazardous nature, high-level nuclear wastes should be removed from the biosphere at the earliest possible time. In the above method, which involves solidification before disposal, early disposal of the waste is not possible without a significant economic sacrifice because of the high rate of heat generation in fresh wastes.

In addition, each operation in the waste-solidification process entails a certain accident potential, and transport of the solidified wastes also carries certain risks. Despite the extensive precautions planned for avoiding such hazards, the possibility of error or accident increases with the amount of handling and movement. Here again, a nuclear chimney may be used, along with the properties of the underground environment, to provide a better solution to this problem. The concept proposed is shown in Fig. 10. The chimney capacities and properties are given as a reasonable example.

To eliminate the need for waste transportation, the disposal chimney would be situated in the immediate vicinity of the nuclear-fuel reprocessing

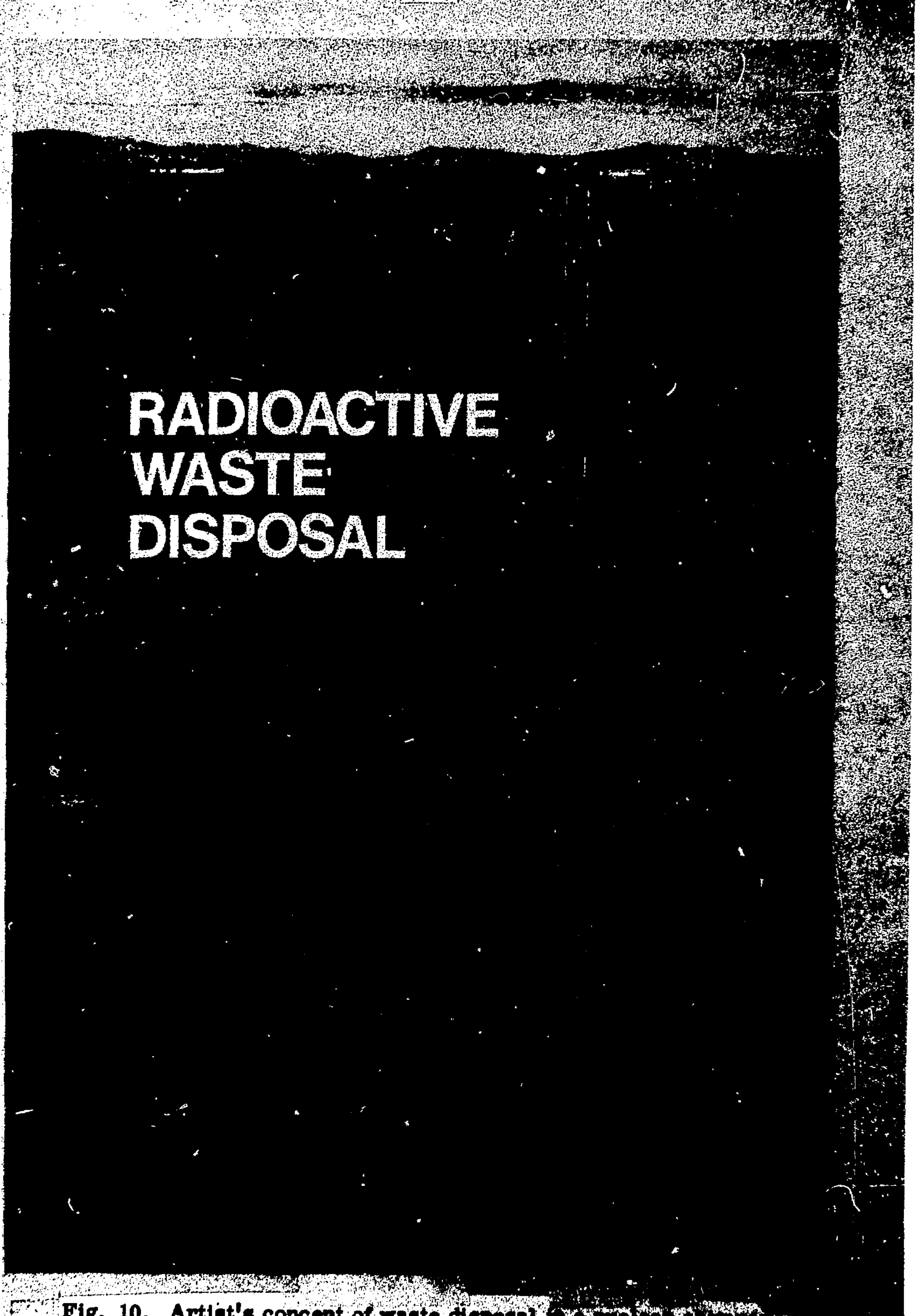


Fig. 10. Artist's concept of waste disposal [illegible]

plant. The chimney would be formed in deep silicate rock having a low permeability to prevent water inflow.

Liquid wastes resulting from the plant operations would be injected into the chimney with minimal treatment. Initially, only fresh, high-level wastes would be injected and allowed to self-boil in the chimney. The resulting vapor would be allowed to escape through a vent to the surface. As soon as enough radioactive material is accumulated to assure continuous boiling, intermediate- and low-level waste would also be added. As the process continues, radioactivity would accumulate and require additional cooling water to prevent the chimney rock from melting. The temperature would be maintained well below the melting point of the rock by regulation of the rate of water addition. At the surface, steam issuing from the vent would be condensed and recirculated to cool the chimney or processed for reuse in the plant. The closed system would ensure no release of contamination to the environment.

When the chimney reaches its capacity (by having its interstitial volume filled with solidified waste), or is no longer needed, waste and water addition would be terminated and the inlet hole would be permanently sealed. When steam and vapor are no longer released, the vent would also be sealed.

In the chimney, the events depicted in Fig. 11 would then occur. For purposes of this description, a 25-yr waste-addition period is assumed. The indicated time spans are based on the plant operating conditions given in Fig. 10 and on heat-flow calculations that will be discussed later.

Fig. 11. Artist's concept of the sequence of events that occur in the chimney when liquid waste is added.

Upon termination of waste and water addition, the chimney temperature would rapidly rise to the melting point of the rock and melting would commence. The chimney rubble would melt first, followed by the surrounding rock. The radioactive material would dissolve in the rock melt. As the molten rock mass increases in size, its surface area would increase, resulting in a higher rate of conductive heat loss to the surrounding rock. Concurrently, the heat production rate of the radioactivity would diminish because of decay. When the rate of heat loss exceeds that of input, the molten rock would begin to cool and solidify. During this period, no environmental hazard could result because the molten rock mass would be essentially immobile (movement to any cooler environment would cause rapid solidification). The heat surrounding the melt would prevent the intrusion of groundwater. The ultimate result would be the permanent incorporation of the waste in an insoluble silicate rock matrix deep underground.

Heat Balance

Consideration of heat generation and transport within the chimney and the surrounding rock is essential to an understanding of the concept.

Figure 12 shows the total thermal power in the processing waste from spent fuel as a function of postirradiation times between 6 mo and 100 yr. A computer code has been used to calculate the heat balance and other conditions in and around the chimney, taking into account such factors as:

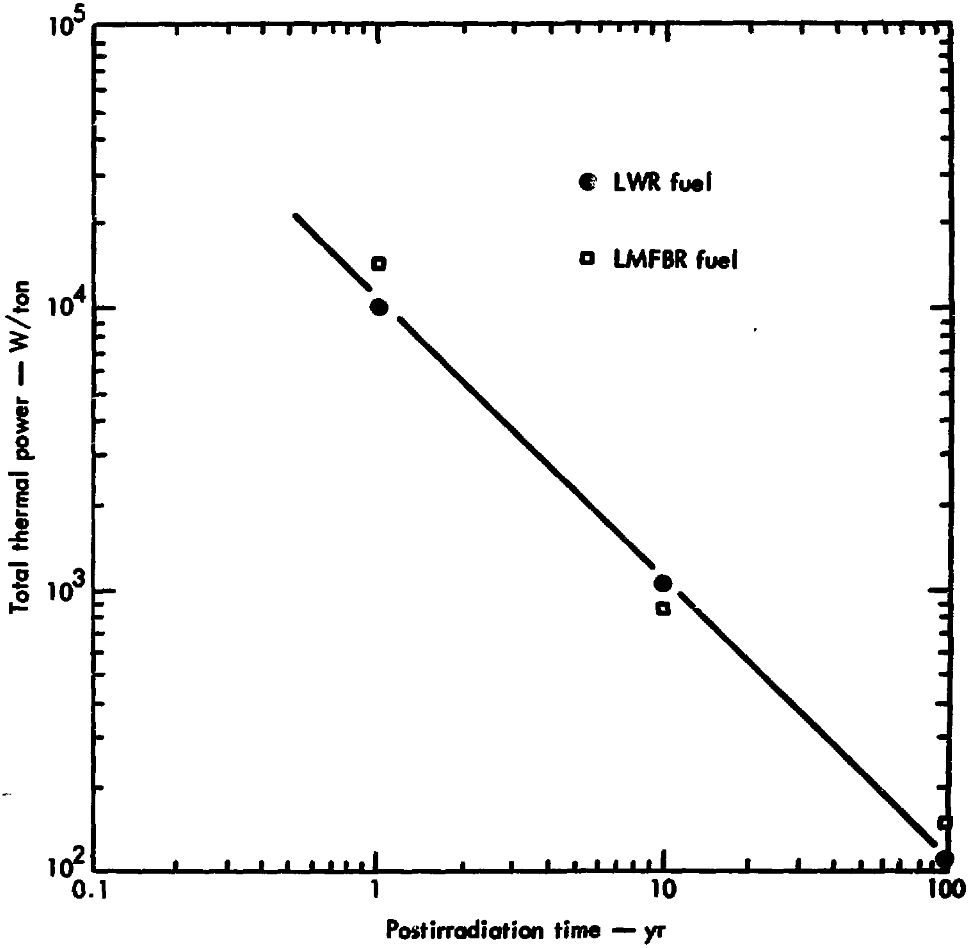


Fig. 12. Total thermal power in the wastes generated by the processing of spent LWR and LMFBR fuel as a function of postirradiation time.

- Rate of waste introduction.
- Thermal power generated by the wastes as a function of time.
- Rock properties such as conductivity, heat capacity, and heat of fusion.

Assumptions used in the calculation are:

- Chimney formed by a 5-kt explosive at a 2000-m depth.
- Age of waste = 0.5 yr (postirradiation time).
- Rate of waste addition = 1500 ton/yr.
- Duration of waste addition = 25 yr.
- Chimney temperature held at 200°C during waste addition.

The results of a typical calculation are given in Figs. 13, 14, and 15. Figure 13 shows the rate of heat production in the chimney peaking at about 67 MW. Figure 14 depicts the required rate of cooling-water addition reaching a maximum of about 1.8 m^3/min at the 25-yr cutoff. Figure 15 shows the radius of the molten rock mass reaching a maximum of 96 m at 90 yr.

Site Considerations

Because transportation of liquid high-level wastes is unacceptable, chimneys would have to be located at the sites of fuel-reprocessing plants. These sites must meet the requirements for both the reprocessing plant and the chimney, together with the required drill holes connecting the surface to the chimney.

A chimney or chimneys could be produced most simply prior to the construction of a plant, but if desired, a plant could be designed and built

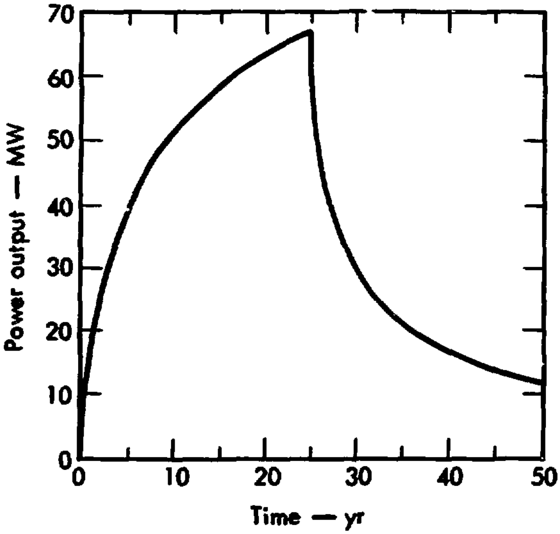


Fig. 13. Power output of waste vs time.

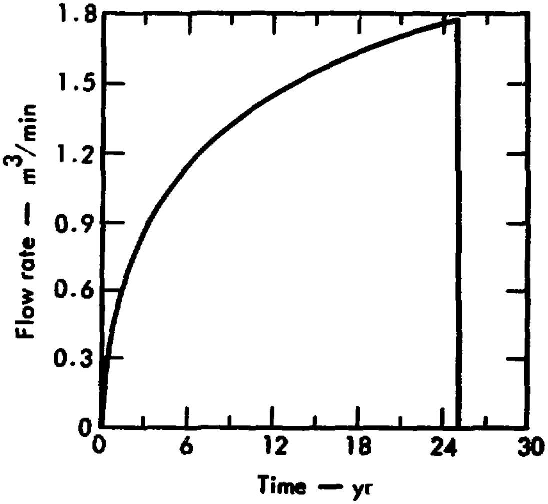


Fig. 14. Required cooling-water rate vs time.

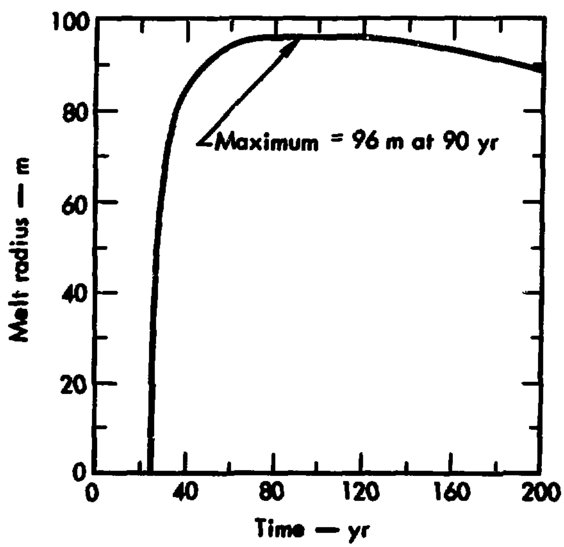


Fig. 15. Melt radius vs time.

to withstand the shock of a nearby 5-kt explosive. If the plant were so constructed, chimneys could be created as needed after construction and connected to the plant by a short pipeline.

Two phases of the operation must be considered in selecting a site with suitable geological and hydrological conditions. These are the waste-addition and rock-melting phases.

In the first phase, the chimney is connected to the surface through drill holes, waste and water are introduced into the chimney, and steam is transported to the surface. In the second phase, after the volatiles are driven out of the chimney, the rock melts and the melt volume increases to some maximum size and then decreases as the melt freezes.

The chimney itself should be placed in rock having a low permeability. This ensures that a negligible amount of water enters the chimney prior to and during the early portion of the first phase and that no radioactivity migrates away from the chimney.

The requirement for the second phase is a low-water-content silicate rock of sufficient dimensions to contain the molten rock at its maximum dimension. This material should contain a negligible amount of carbonate rock in order to avoid the generation of CO_2 during the melting phase. As soon as the rock starts to melt, the radioactive materials are dissolved in the melt and are soon surrounded by molten rock. The peripheral melt zone will contain little or no radioactivity because of the high viscosity of silicate rocks near their melting point.

In order to provide further assurance that the waste is permanently isolated from the biosphere or from any water that might find its way to the surface, the chimney would be constructed below a considerable thickness of impermeable rock and at a depth greater than 2000 ft (~600 m).

During the first phase, when the drill holes are transporting water and steam between the chimney and the surface, care must be taken to avoid the introduction of radioactivity into rock zones containing mobile water. The system (chimney and holes) would be operated at a pressure less than that in any water-bearing zones, except perhaps those within a few hundred feet of the surface. Any leak in the well casings below this depth or in the chimney would therefore be into the system rather than out of it. To avoid contamination in the shallow zone, a site should be selected with no important aquifers within several hundred feet of the surface, and preferably none at all.

The required geologic and hydrologic conditions for a site are commonly available and could be confirmed with minimum exploration costs.

Operational Considerations

Figure 16 gives a generalized plan for an integrated nuclear-fuel reprocessing and waste-disposal facility. As soon as the chimney becomes self-boiling, it would accept intermediate- and low-level waste, as well as high-level waste. This waste plus the water from the condenser would be used to supply cooling water to the chimney. The principal function of the condenser would be to circulate enough water into the chimney, and steam out, to keep the chimney well below the melting point of the rock during the operation of the facility.

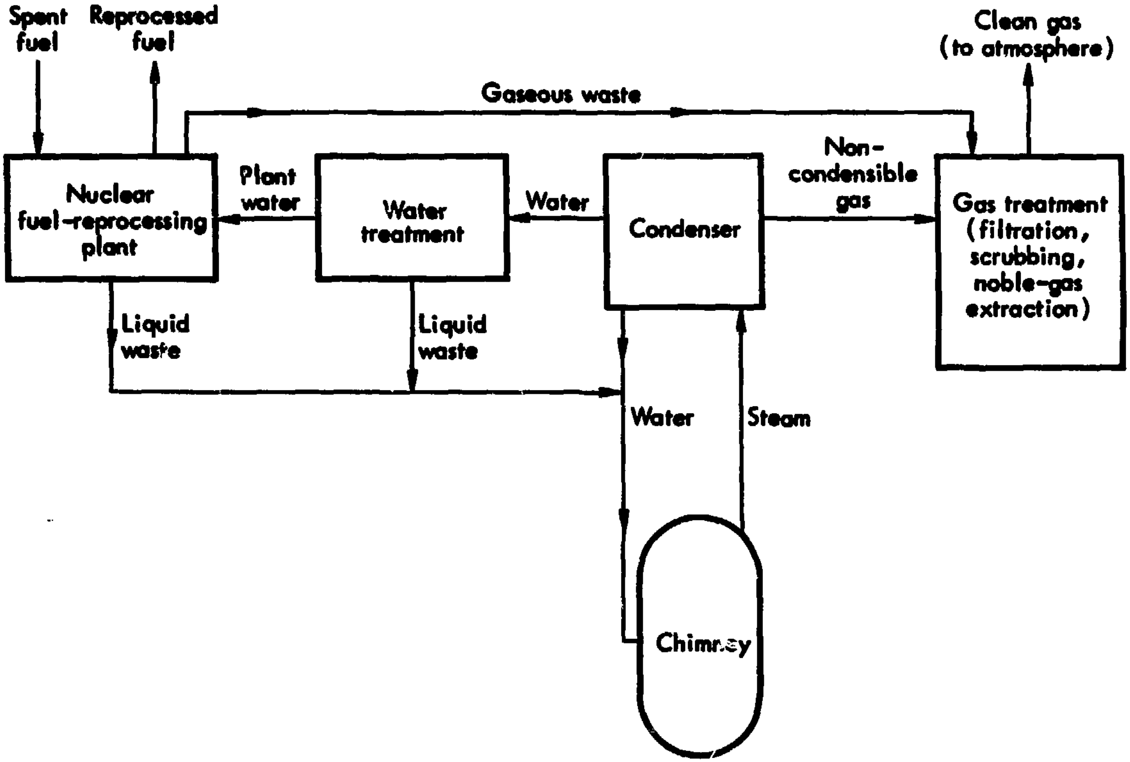


Fig. 16. Generalized flow diagram for an integrated nuclear-fuel reprocessing and waste-disposal facility.

Economic and Environmental Advantages

The cost of high-level waste disposal using present state-of-the-art methods is estimated to be between \$7,600 and \$12,800 per ton of fuel processed. This does not include the cost of medium- and low-level waste disposal. The method proposed here is estimated to cost about \$2,000 per ton, including medium- and low-level wastes. By the year 2000, when an estimated 4.4×10^{12} kWh/yr will be generated by nuclear reactors, use of this method could result in an annual savings of several hundred million dollars.

The major concern regarding nuclear waste disposal, however, is the avoidance of environmental pollution. In the concept presented, waste storage and processing are minimized, and the need for transport of the wastes is eliminated. In addition, this concept offers several other environmental advantages:

- Prompt elimination of the radioactive waste from the biosphere.
- Permanent binding of the radioactive waste in a rock matrix deep underground, giving assurance of its permanent elimination from man's environment.
- Provision of a safe method of permanently disposing of intermediate- and low-level wastes.

CONCLUSION

The use of nuclear chimneys for the in situ, high-pressure, high-temperature recovery of copper from sulfide ores and the in situ incorporation of nuclear-fuel reprocessing waste in molten silicate rock are two examples of future uses of the underground environment. In both examples, properties of the underground environment are used to advantage in processes that would be impractical above ground.

REFERENCES

1. E.G. Teller, W.T. Talley, G.H. Higgins, and G.W. Johnson, The Constructive Use of Nuclear Explosives (McGraw-Hill Book Co., New York, 1968), Chap. 4.
2. T.R. Butkovich and A.E. Lewis, Aids for Estimating Effects of Underground Nuclear Explosions, Lawrence Livermore Laboratory, Rept. UCRL-50929 (1970).
3. A.E. Lewis, "Chemical Mining of Primary Copper Ores by Use of Nuclear Technology," in Proceedings of Symposium on Engineering with Nuclear Explosives, Vol. II (American Nuclear Society, CONF-700101, 1970), pp. 909-917. Available from the National Technical Information Center, National Bureau of Standards, U.S. Department of Commerce, Springfield, Virginia 22151.
4. A.E. Lewis, "Chemical Mining of Primary Copper Sulfide Ore Using High-Pressure Oxygen in a Flooded Nuclear Chimney," presented at the 1971 Idea Conference, New Mexico Institute of Mining and Technology, Socorro, New Mexico; Lawrence Livermore Laboratory, Rept. UCRL-73109 Abstract (1971).
5. R.L. Braun and A.E. Lewis, "Chemical Mining of Primary Copper Sulfide Ores—O_2 Distribution, Fluid Circulation, and Rate of Copper Recovery," presented at the 1971 Idea Conference, New Mexico Institute of Mining and Technology, Socorro, New Mexico; Lawrence Livermore Laboratory, Rept. UCRL-73110 Abstract (1971).

6. J. J. Cohen, A. E. Lewis, and R. L. Braun, Use of a Deep Nuclear Chimney for the In Situ Incorporation of Nuclear-Fuel Reprocessing Waste in Molten Silicate Rock, Lawrence Livermore Laboratory, Rept. UCRL-51044 (1971).

7. J. J. Cohen, A. E. Lewis, and R. L. Braun, In Situ Incorporation of Nuclear Waste in Deep Molten Silicate Rock, Lawrence Livermore Laboratory, Rept. UCRL-72848 Rev. 1 (to be published).

8. "Siting of Commercial Fuel Reprocessing Plants and Related Waste Management Facilities," statement of Proposed Policy 10 CFR, Part 50, Fed. Reg. 35:222 (1970), p. 17530.