

REACTOR WASTE UTILIZATION
IN SEWAGE SLUDGE TREATMENT
H. D. Sivinski

I. INTRODUCTION

Land application of sewage sludge has received some attention in the past, but present conditions of growing urban populations, energy shortages, and the world food problems are bringing renewed and vigorous interest to this activity. The utility of sludge and its nutrients in the form of nitrates, phosphates, and other organic and inorganic compounds has been amply demonstrated in research reported on at various symposia and in the literature [1], [17]. The very fact that it will become a major asset in attacking the problems of fertilizer shortages, land reclamation, and increased food production means that adequate safeguards for its use must be developed. Agronomists are vigorously pursuing the problem of plant uptake of heavy metals. At a recent agronomy symposium in Chicago [2], some 47 papers were given in six sessions on sludge. The majority of these dealt with the heavy metals problems. The problem of pathogen reduction was less well represented since it was felt by many authors that public health and environmental protection agencies were largely responsible for research activity in this area.

A variety of methods for reduction or elimination of such pathogens remaining in sludge after aerobic or anaerobic digestion can be used. Heat treatments such as pasteurization, chemical treatments such as liming and ionizing radiation with both electrons and gamma rays have been used. Extended storage time will also inactivate a large number of pathogens. The research described in this paper deals with inactivation of sewage pathogens by means of heat and ionizing radiation (gamma) applied singly and thermoradiation (TR), heat and ionizing radiation applied simultaneously.

II. HEAT ACCELERATED RADIATION EFFECTS

Much of the early work at Sandia Laboratories in heat accelerated radiation effects was done with dry *Bacillus subtilis* var. *niger* spores by M. C. Reynolds and co-workers. It was this work which indicated that synergism could be obtained when properly chosen combinations of heat and ionizing radiation were simultaneously applied [3], [4], and [5]. Synergism is defined as that part of an inactivation result greater than the additive effects of heat alone and of ionizing radiation at ambient temperature as shown in Figure 1. Figure 2 shows results obtained at 105°C with a soil spore and indicates the degree of synergism obtained.

The work was subsequently expanded to look at many temperatures and dose rates [6], such as shown in Figure 3 and with other soil spores [7], Figure 4. Work was done with bacteriophage and virus [8], [9], Figures 5 and 6, which indicated these systems also responded synergistically when proper combinations of heat and ionizing radiation were applied simultaneously. Vegetative systems such as *Escherichia coli* also show a

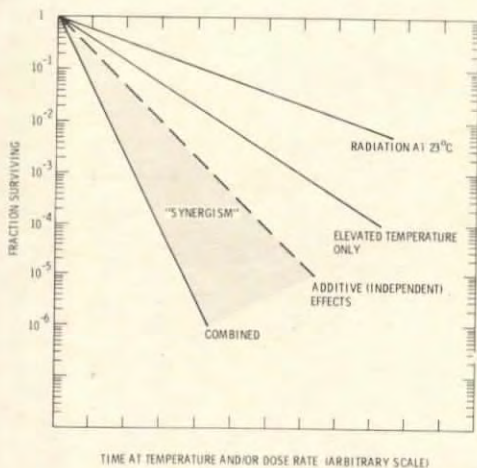


Figure 1. Illustration of synergistic response to simultaneous application of heat and ionizing radiation.

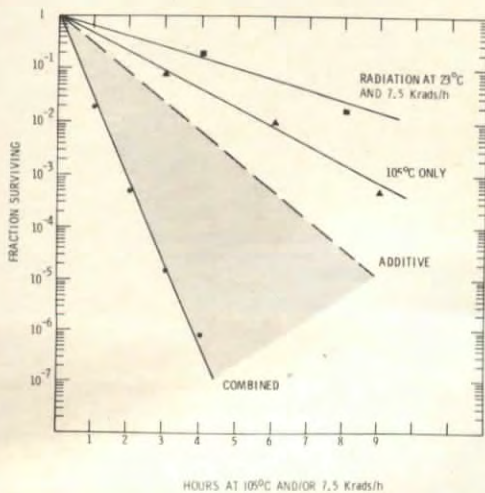


Figure 2. Inactivation of dry *Bacillus subtilis* var. *niger* spores at a dose rate of 7.5 krads/h and separately and simultaneously.

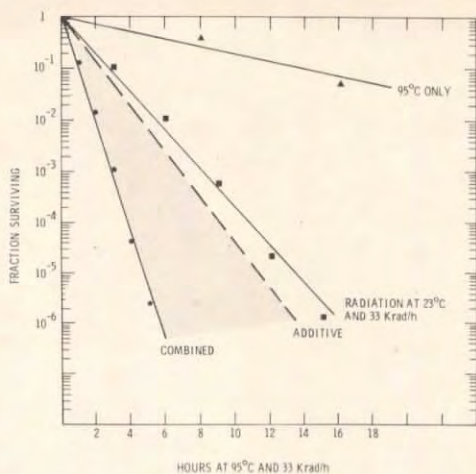


Figure 3. Inactivation of dry *Bacillus subtilis* var. *niger* spores at a dose rate of 33 krad/h and 95°C separately and simultaneously.

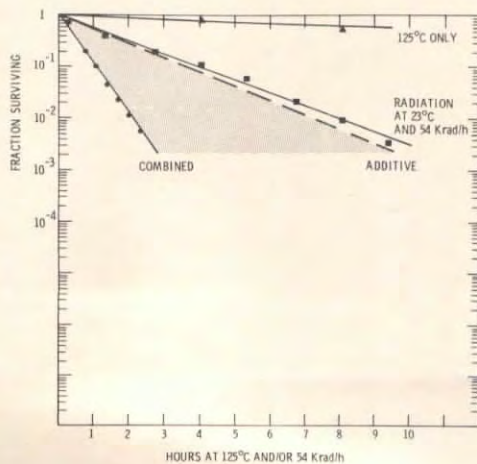


Figure 4. Inactivation of dry, unidentified heat resistant soil spores at a dose rate of 54 krad/h and 125°C separately and simultaneously.

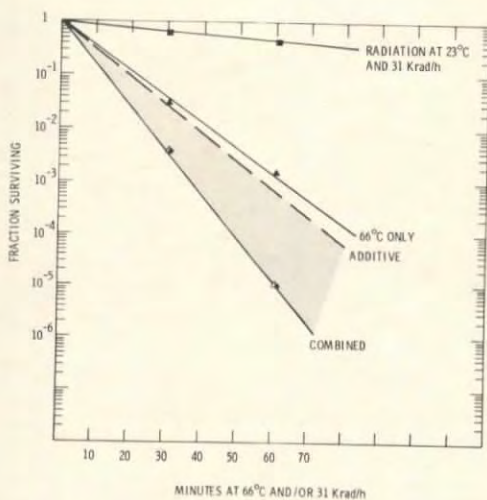


Figure 5. Inactivation of T4 bacteriophage in broth at 31 krad/h and 66°C separately and simultaneously.

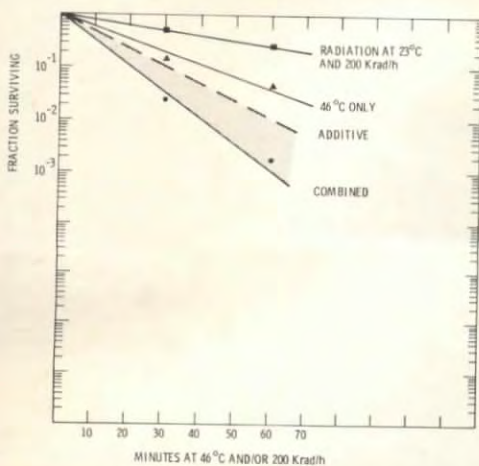


Figure 6. Inactivation of poliovirus (LSC) in MEM complete at 200 krad/h and 46°C separately and simultaneously.

synergistic response [10], Figure 7. These synergistic phenomena have been extensively modeled by Brannen [11], [12], [13], and Dugan and Trujillo [14], [15], all of Sandia Laboratories. The work done since 1968 supports the conclusion that, when properly chosen, combinations of heat and radiation act synergistically to inactivate biological systems [16]. Because one has to look at heat and at radiation separately and then at the combination in order to determine synergistic effects, a great store of such data on the response of certain biological systems to these treatments has been developed at Sandia. Analysis of these data indicate that:

- i. Treatment time at temperature can be reduced with ionizing radiation, or
- ii. Radiation dose can be reduced significantly by adding heat.
- iii. When thermoradiation combinations are properly chosen, synergism is observed with aerobic and anaerobic spores, vegetative cells, viruses, and enzymes.
- iv. Such synergism can result in up to 30-fold reduction in treatment times.

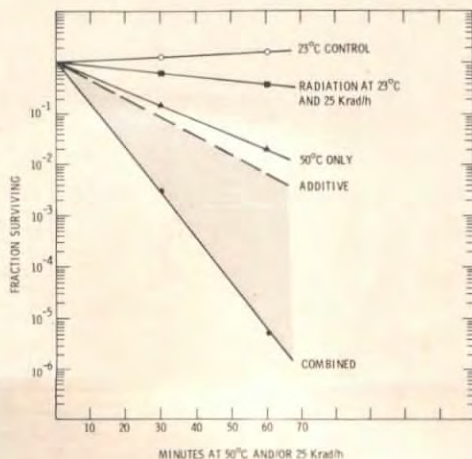


Figure 7. Inactivation of *Escherichia coli* in broth at 25 krad/h and 50°C separately and simultaneously.

III. A CONCEPT AS A SOLUTION

In our work with thermoradiation over the past six years, we at Sandia have been aware of the reduced thermal and source costs for the process. We have studied the food sterilization problem [18], the medical and pharmaceutical applications problem [16], blood sterilization [19], etc. It was the projected use of large numbers of curies of source material for such programs which indicated that the use of Cesium-137 recovered from radioactive waste would be necessary. Although configured poorly for use as sources, the capsules of Cesium-137 from existing radioactive waste resources are now available and have been advertised in the Federal Register at a cost of 10¢/curie. This resource, coupled to the future generation of large amounts of reactor waste product, provides rationale and credibility to the plan for use of high-level radiation in waste treatment. The Waste Resources Utilization Program was organized at Sandia in 1973 to study and do research in the most promising of such waste treatment concepts. The major work is an attempt to constructively couple two major environmental programs, disposition of human and nuclear waste materials, toward a mutually beneficial solution, the utilization of nuclear reactor waste products, Cesium-137 and Strontium-90, in the thermoradiation treatment of municipal sludge. The sludge treated in this way would provide a valuable material for agriculture and land reclamation, free of the serious potential health hazards associated with conventional methods of land disposal. Heat required for treatment could be supplied by using the reactor waste product, Strontium-90, as a heat source or by using the methane generated in the anaerobic digestion stage of conventional treatment. The cost per curie of the radiation sources would be minimized by using large quantities of reactor waste products.

IV. THE SANDIA LABORATORIES WASTE RESOURCES UTILIZATION PROGRAM

The thermoradiation studies over the last few years at Sandia show significant promise due to the synergistic inactivation exhibited in all biological systems studied. The research has more recently been pointed toward investigation of thermoradiation for disinfection of sewage sludge using nuclear reactor waste fission products [17]. If this technology can be successfully transferred to sludges, it could provide a cost-effective alternative for sludge disinfection and treatment and for the beneficial utilization of intermediate-life radioactive isotopes [20]. The research is directed toward determination of the lowest cost effective temperature and radiation dose to meet the minimum level of disinfection of digested sludges that would be accomplished by pasteurization treatment at 70°C for one-half hour.

Biological Effects

The biological research is currently concentrated in three major areas: bacteria, viruses, and parasites. Both 208,000 Ci Cesium-137 and 87,000 Ci Cobalt-60 sources have been used. Tests have shown that the Relative Biological Effectiveness (RBE) for pathogens and parasites is the same for both sources.

Bacteria - The bacterial systems being used to study thermoradiation inactivation in sludge are fecal streptococcus and fecal coliforms. The sludge is now being withdrawn from the first of two anaerobic digesters used in series by the Liquid Waste Division of the City of Albuquerque. The anaerobically partially digested sludge averages 8% solids. It is strained, blended and then treated. A typical result for thermoradiation treatment of fecal coliform is shown in Figure 8. The synergism is not large but it is important to note the tailing of the 57°C heat curve. The addition of 60 krad in 12 minutes essentially inactivates the coliforms present.

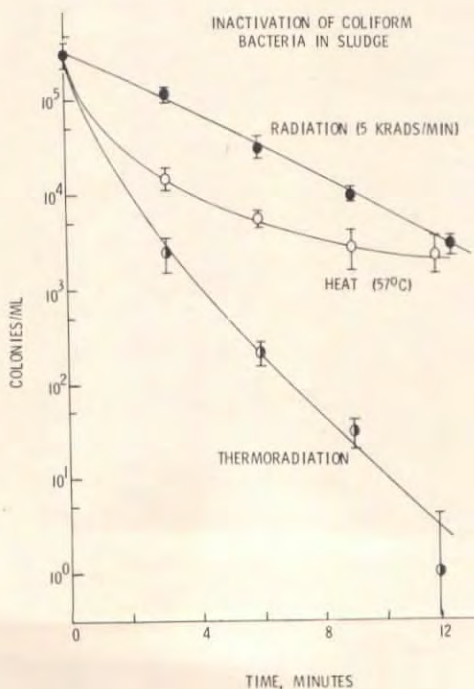


Figure 8. Inactivation curves for heat, radiation, and thermoradiation treatment of coliforms in sludge.

The same type of problem appears with fecal strep in sludge, Figure 9, i.e., the heat treatment results in tailing. Even though the fecal strep appears some 8 times more radiation resistant than the coliforms, addition of heat at 57°C provides a dramatic increase in inactivation as one approaches sterility.

Research at various temperatures and dose rates is needed to determine optimal thermoradiation inactivation rates for these bacteria. Some work has already been done [21] and the test series will continue in both laboratory bench tests as well as the dynamic liter quantity flow through system described later.

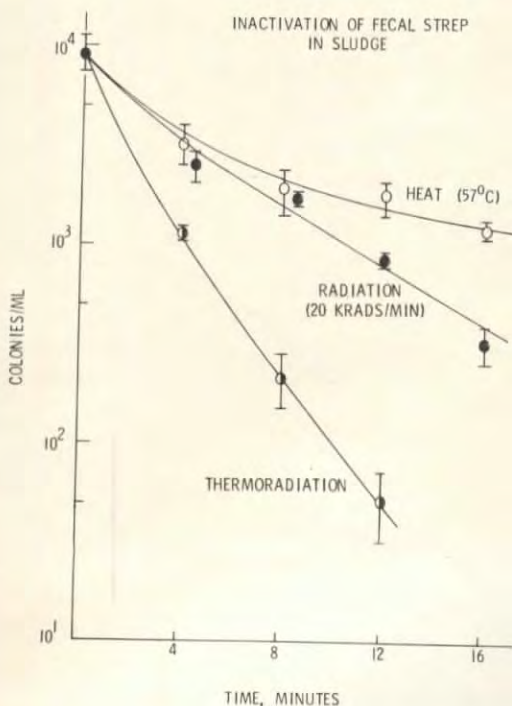


Figure 9. Inactivation curves for heat, radiation, and thermo-radiation treatment of fecal streptococci in sludge.

Another area being investigated is the mutagenic effect of sublethal dosing on bacteria. Very preliminary data [22] suggest that thermoradiation combinations may produce fewer mutants among survivors than radiation at room temperature. The work strives to assess the effect of heat, radiation, and thermoradiation in production of auxotrophic mutations in dry *Bacillus subtilis* var. *niger* spores (Figure 10). Whether conclusions drawn from this study may be applicable to other classes is unknown at this time. The percent mutants among survivors may be as high as 15% for radiation while approaching natural background level for thermoradiation cycles. Although it is too early to know how generally valid such behavior might be, these results may offer yet another potential benefit for use of a thermoradiation cycle in bacterial inactivation.

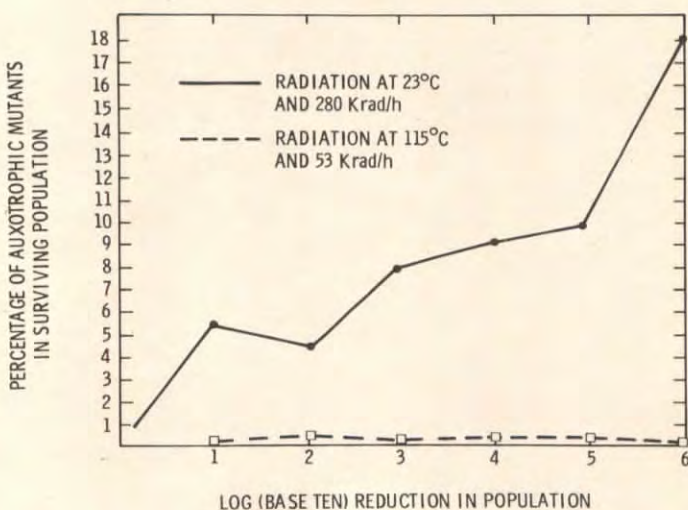


Figure 10. Auxotrophic mutants induced in populations of dry *Bacillus subtilis* var. *niger* spores by radiation at ambient and elevated temperature.

Viruses - Viruses are generally known to be quite resistant to radiation, their resistance depending upon the amount of dominant nucleic acid present. The D_{37} ranges from about 200 krad up to 1000 krad [23]. The radio-sensitivity of viruses is known to depend upon the nature of the media in which the irradiation takes place (e.g., [24]). Similarly, with heat, the rate of inactivation depends upon strain, its past age history, and the nature of the suspending medium. Generally, enteroviruses are less heat resistant than rhinoviruses [25], while being of major concern in sewage sludge treatment. One typically expects fairly rapid inactivation of enteroviruses at 50°C, although this definitely depends upon the nature of the suspension medium and strain of virus. Survival of some strains of poliovirus has been reported at 50°C [26] for many hours, while hepatitis B is known to survive up to 4 hours at 60°C in unknown titer [25].

Our work to date has centered in two areas: Studies with T4 bacteriophage (as an early model system) and with poliovirus. Figures 5 and 6 presented earlier are indicative of results. The temperature region in which T4 phage exhibits a synergistic response at 30.6 krad/hour is about 55-70°C [27]. At higher dose rates, this interval is somewhat larger. Two strains of poliovirus are being examined. The data in Figure 6 are for the LSC strain. Here, the temperature is 46°C at a dose rate of 200 krad/hour is insufficient for this strain of poliovirus, it is relatively close to a good combination since heat alone at 49°C will inactivate about 3 logs of virus in 5 minutes of treatment. Other data suggest that even slightly higher temperatures will be desirable [26]. Generally, it is anticipated that ranges comparable to those needed for fecal strep will be adequate for virus inactivation. The case of viruses treated in sludge.

Ascaris - The parasite system of choice for TR inactivation of ova in sludge was Ascaris lumbricoides. It has been reported to be radiation resistant [28] and is ubiquitous. The research program will be complete in July 1975 and a preliminary report is in press [29]. The protocols were developed for water initially because of handling, embryonation, and counting problems. The work has now been essentially completed in sludge. As a point of interest, sludge was only slightly more protective for the heat and radiation tests than was water. The more significant findings are listed below:

- i. In the heat treatment work, 51°C was established as the temperature at which heat effects begin to become observable. No inhibition is apparent for the first 10 minutes at this temperature, but inhibition of embryonation is complete after 50 minutes. There was no inhibition of embryonation at 47°C up to 2 hours of exposure, while nearly complete inhibition was obtained in 6 minutes at 55°C.
- ii. Radiation at ~21°C using a Cobalt-60 source and a dose rate of ~36 krad/minute cause a one log (90%) drop in percent embryonated at 45-50 krad. The second log drop occurred at 65-70 krad and complete inhibition was observed at ~95 krad.

- iii. The thermoradiation tests were run at 47°C, a temperature at which heat alone shows no effects after 2 hours. At the dose rate of 24 krad/minute synergy was quite apparent. At ambient temperature the lethal dose required to prevent embryonation was 95 krad. At 47°C the lethal dose was 40 krad or only 42% of the dose needed at ambient temperature. The data enumerated above are summarized in Figure 11.

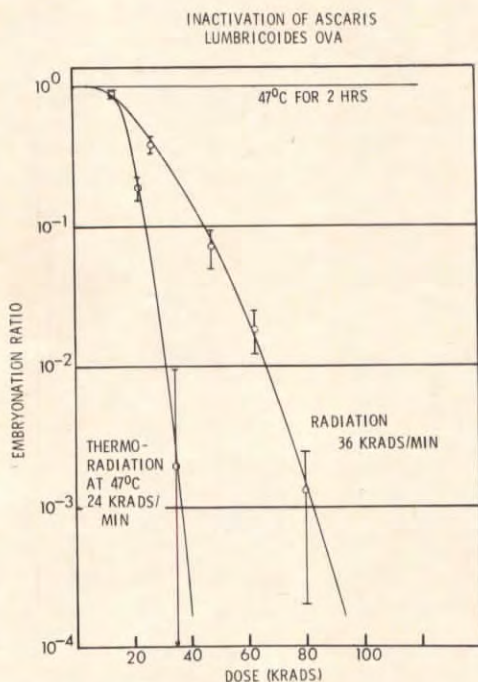


Figure 11. Comparison of heat, radiation, and thermoradiation effects on the embryonation of *Ascaris lumbricoides* ova.

Chemical and Physical Effects

Ionizing radiation has been observed [30], [31], to enhance sludge settling rates. We have found a significant increase for anaerobically digested sludge treated with several hundred kilorads of Cobalt-60 gamma rays [32]. Figure 12 shows a typical improvement versus dose curve. It is seen that the enhancement in initial settling rate begins with doses as low as 50 krad; a plateau is approached at higher doses. The experiment requires proper controls, since the settling rate and the improvement depend on so many variables: Percent settled, solids content, previous treatment of the sludge (agitation, chemicals), and composition. The sludge used in determining Figure 12 was diluted with primary digester supernatant to about 4% solids.

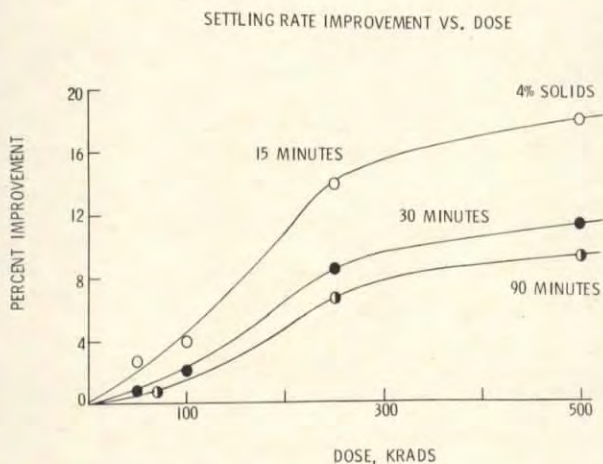


Figure 12. Improvement in sludge settling rate due to 400 krad Cobalt-60 irradiation. Solids content approximately 4%.

While heat affects settling rate only slightly (60°C), it can be seen in Figure 13 that thermoradiation treatment appears to act synergistically, i.e., the improvement noted for thermoradiation is greater than that for radiation alone (heat alone is negligible at longer times). These results are preliminary, and studies are currently under way on the radiation-chemical aspects of this phenomenon. We are also beginning filterability measurements in terms of heat, radiation, and thermoradiation.

In line with radiation-chemical aspects, we have been measuring chemical oxygen demand (COD) changes induced by radiation, both in settled sludge and in "post-settling" supernatant. We have observed radiation-induced increases in supernatant COD, implying that partial breakdown of organics (and therefore increased solubility) or change effects are occurring.

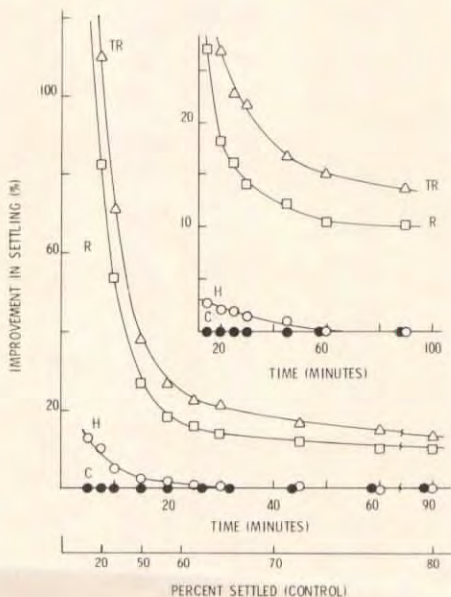


Figure 13. Improvement in sludge settling for heat (H, 60°C), radiation (R, 400 krad), and thermoradiation (TR, 400 krad, 60°C). Solids content approximately 4%.

We are beginning measurements on other physical and chemical changes induced by ionizing radiation in sewage sludge (odor modification, refractory modification, etc.), but results are qualitative at best.

Concurrent with the research cited above, Sandia is also performing systems design and realistic cost benefit analyses of the thermoradiation studies were made previously [17] to determine feasibility but as more is learned about the process and as costs change, further studies are necessary. Inherent in such analyses is work in prediction of radio-isotope requirements for the foreseeable future and initiation of studies of the options for their procurement and safe use. The development of programs leading to public acceptance of radiation treatment of wastes is also of concern.

The full scale facility design concept currently under consideration uses a single pass flow through system, Figure 14. The research investigating such a system is currently being done on a laboratory scale

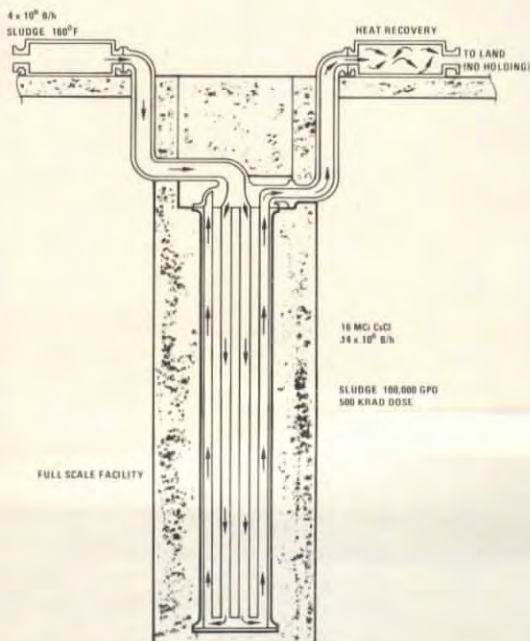


Figure 14. Full scale Cesium Irradiation Facility

recirculation system with dynamic sampling. A larger dynamic liter quantity single pass flow through system with continuous sampling capability has been fabricated and assembled (Figure 15). The sludge will begin to be treated in barrel quantities on this system in May 1975. This research will lead to the development of a 50,000 gallon per day (gpd) pilot plant which should provide design parameters for full scale facilities by 1979. The timetable for milestone achievement to bring such a facility on line is shown in Figure 16.

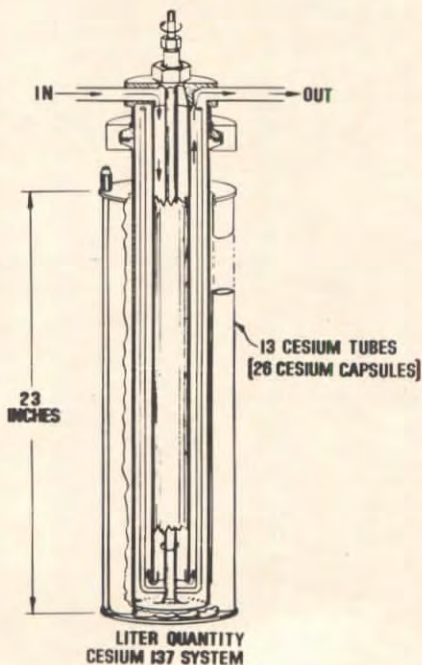


Figure 15. Liter quantity Cesium-137 system

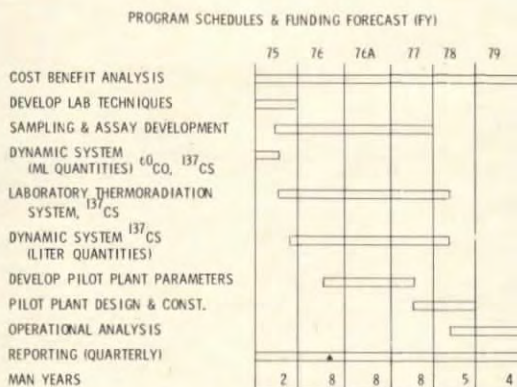


Figure 16. Timetable for milestone achievement to complete the pilot plant studies

Far reaching benefits may arise from these studies. The successful transfer of the thermoradiation technology with beneficial use of nuclear reactor waste in the solution of the land application of sludge problems can lead to increased food production, protection of the environment, and enhancement of the quality of life.

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