

even if we were to burn this gas for power, the total reserves of gas in West Pakistan, as discovered up to date, although there may be more, would be equivalent to about 365 million tons of coal, which is about two years' consumption of the United Kingdom at the present rate. We do not have very large reserves of gas as is commonly believed. In any case, I for one in Pakistan can never advise my Government in such a way that by the time my son, who is 4 years old, grows up to my age and becomes a petrochemical or a chemical engineer and asks "where is the gas of Pakistan", he is told "your daddy has burnt it for power generation". We do not want to be in that situation.

In East Pakistan unfortunately there is no

hydro potential at all. We have about 120 MW for a population of about 51 million, an area peopled with the highest density of population in the world. There is no major discovery of gas comparable to the big deposits found in West Pakistan; there is no oil and no coal. In East Pakistan we are importing about 80 000 tons of coal per month from South Africa, China, Poland, India and everywhere to keep the economy going. We have to break the ice somewhere as far as power is concerned and in East Pakistan we have already decided to start with a modest nuclear power programme. We are thinking of establishing a water-moderated 50 MW reactor in East Pakistan to be operative some time in 1967-68 and about a 100 MW reactor in West Pakistan in the Karachi area where the load factor is fairly high.

HIGH-LEVEL RADIOACTIVE WASTES

Methods of Treatment and Storage Discussed at IAEA Symposium

Management of radioactive wastes produced in reactor operations is one of the important problems in the atomic energy industry, and since the expansion of the industry involves an increase in the volume of the wastes produced, it is also a problem of growing magnitude. While the hazards from the radioactive by-products of atomic operations must be reduced to the minimum possible in a given state of technology, there should be a simultaneous effort to ensure that the measures to achieve safety are sufficiently practical and economical for application on a large scale.

Safety, of course, must remain the primary consideration, and attempts are being made at atomic energy centres all over the world to devise the safest and most effective methods of dealing with radioactive wastes. From the beginning, the International Atomic Energy Agency has considered it one of its main tasks to stimulate and co-ordinate this effort; in fact, work in this field has constituted one of the Agency's most comprehensive programmes. An important part of this programme has been the organization of scientific meetings to discuss different aspects of radioactive waste management.

Certain aspects of the subject have been, and are being, examined by small groups of experts convened by the Agency, while common scientific and technical problems are discussed at larger meetings. In organizing these discussions, the Agency has divided the problems into two broad groups: those

relating to wastes of large volume but with a low level of activity and those of a relatively small volume but with a very high level of activity. Although the distinction between high and low levels of activity is largely a matter of convenient definition, current usage of the two terms puts the wastes from the operation of small research reactors or from radio-isotope applications into the low-level category, while wastes generated from the reprocessing of spent reactor fuel - such as from nuclear power plants - are placed in the high-level category. Apart from being intensely radioactive, the latter, in many cases, retain their radioactivity for very long periods, often for thousands or hundreds of thousands of years. It is the combination of these two characteristics which makes these substances especially difficult to deal with. If the level of activity is very low, it may be possible to disperse the wastes into the environment, but substances with a high level of activity must be kept isolated. If the radioactive products are short-lived, the isolation need only be temporary, but high-level wastes with a long radioactive life may have to be isolated from the human environment for thousands or more years. In other words, they must be stored in such a way that they cannot escape into the environment so long as they remain a potential source of radiation hazards; and before storing them, it is usually necessary to treat the wastes by various chemical processes so as to make them suitable for storage.

This is a task of considerable complexity; in fact, the treatment and storage of high-level radioactive wastes constitute the biggest single problem in atomic waste management. And the magnitude of the problem can be gauged from the fact that in the United States alone more than 50 million gallons of high-level wastes are now stored in underground tanks. If one takes into account the large nuclear programmes of certain other countries as well as the likely expansion of the nuclear power industry in many parts of the world, one can arrive at a rough estimate of the vast quantities of high-level wastes that will have to be safely stored in the years to come.

International Symposium in Vienna

Methods of treatment and storage of high-level wastes were discussed at an international symposium organized by the Agency in Vienna from 8 to 12 October 1962. One hundred and thirty scientists from 19 countries and from the European Nuclear Energy Agency and EURATOM attended the meeting.

Apart from a general review of problems and current practices, the work of the symposium was divided into six sections, under the following headings: (1) concentration and storage, (2) solidification and fixation of liquids by calcination, (3) solidification and fixation by vitrification, (4) solidification and fixation by miscellaneous methods, (5) treatment of solid wastes, and (6) shipment of gross quantities of radionuclides. The different sessions were presided over by the following scientists: P. Dejonghe (Belgium), F. Duhamel (France), E. Glueckauf (United Kingdom), L. P. Hatch (USA), B. Kolychev (USSR), and D. Pearce (IAEA).

Opening the symposium, Mr. Pierre Balligand, the Agency's Deputy Director General in charge of Technical Operations, pointed out that this was the first time that the Agency was holding a scientific meeting specifically to discuss problems connected with the management of wastes from the chemical reprocessing of nuclear fuels. He added that it had been considered appropriate to organize this meeting at the present time in view of the fact that data on the design, construction and operation of several demonstration facilities for the treatment and storage of high-level wastes had only recently become available. The Agency's Director of the Division of Health, Safety and Waste Disposal, Dr. Dennis Pearce, said that information exchanged at the symposium would be useful not only to scientists from the advanced countries who were already tackling the problems involved, but also to scientists from countries where these problems were likely to arise in the near future.

The papers presented at the meeting described the present techniques of high-level waste management in the atomically advanced countries, as well

as the direction of research and development aimed at finding better methods of handling the problem. A significant fact that emerged from the information disclosed at the meeting was a broad similarity of approach on the part of all countries dealing with high-level radioactive wastes. It was clear that in almost all cases such wastes are now stored in underground tanks - mostly of stainless steel and concrete. Since, however, the radioactive life of some of the wastes may outlast the life of the most sturdily built tanks, there has been - as was evident from the discussions at the symposium - a general search for a suitable method of solidifying the wastes or fixing them in some solid materials, which would prevent their dispersion into the environment even after the decay of the containers. It was generally recognized that the ultimate solution of the problem of high-level wastes lay in immobilizing them in such a way that they might be permanently isolated from the human environment.

For solidification and fixation, two principal approaches were considered: (1) calcination, i.e. heating the liquid wastes so as to make them form solid oxides, and (2) vitrification, i.e. fixation and incorporation of the wastes in glass or glass-like substances. It was generally felt that while the formation of solid oxides would represent a considerable advance towards immobilizing the wastes, it would not be a permanent solution because under certain conditions the radioactive substances incorporated in the oxides could free themselves and thus possibly find their way into the general environment. Fixation in glass, on the other hand, would be virtually permanent, and a number of reports were presented at the symposium on current efforts to develop this method.

Present Practices and Plans

In a paper on the concentration and storage of high-level wastes from the first stages of the UK civil nuclear power programme, D. W. Clelland (UK) explained that two forms of highly radioactive waste arise during the processing of irradiated reactor fuel. The first is a solid waste, produced during the removal of the can from the fuel, which is stored in concrete silos and presents "no undue technical problems". The second is a liquid waste which is produced when the irradiated fuel, after removal from the can, is dissolved in nitric acid and subjected to a process for the separation of uranium and plutonium from the fission products. The waste contains the fission products after the uranium and plutonium have been extracted.

Mr. Clelland said that the present UK system for dealing with highly active liquid wastes involved, in the first place, a concentration of the acidic liquid by evaporation to a small bulk, followed by storage in stainless steel tanks which were housed in thick-

walled concrete cells. The cell walls were lined with stainless steel to form a secondary containment, and the heat generated from radioactive decay of the fission products was removed by circulating water through cooling coils built into the storage tanks.

Describing the concentration and storage of fission products at the Marcoule plutonium extraction plant, M. Chambon and J. Rodier (France) stated that the very highly active effluents of the plant were first concentrated by evaporation at atmospheric pressure. The concentrated solution of fission products was then led, by gravity, into stainless steel tanks with a useful capacity of 60 cubic meters. The wastes were cooled by closed-circuit water circulation in the pool in which each tank was immersed and within internal coils. A permanent check was maintained on the volume and temperature of the stored liquids, on pressure in the storage tanks and on the activity of the cooling circuits.

Current waste storage practices in the United States were described in a paper by B. F. Campbell, E. Doud and R. E. Tomlinson (USA). They said that for storage purposes high-level wastes were "segregated into two categories, depending on relative fission product content". One category comprised what was described as "self-heating" wastes, i. e. those in which enough heat was generated by radioactive decay of fission products to heat the liquid to boiling, while the other category comprised what was described as "non-heating" wastes, i. e. the less active wastes, which nevertheless contained enough radioactive materials to require storage. For the self-heating wastes, it was necessary to make special provisions for the safe dissipation of the heat generated.

Regarding storage facilities, the authors said that a wide variety of tank designs had been used, depending on variations in volumes and compositions of the wastes, in environmental conditions and in engineering judgment factors. The capacity of most of the tanks ranged from 300 000 to 1 330 000 gallons; they were about 50 to 85 feet in diameter and had a usable depth ranging from 18 to 32.5 feet. The tanks were generally covered with seven to ten feet of earth, as a radiation shield. Giving an example, the authors described the type of tank used at Hanford for the storage of self-heating wastes - a reinforced concrete tank lined with carbon steel to above the liquid level, but without any lining on the dome.

In another paper, R. E. Tomlinson described the high-level waste management programme at the Hanford plant of the USAEC. The objectives of the programme, he said, were to control and minimize the hazards, recover by-products as requested by the AEC, and reduce the cost of waste management. It had been concluded that the best approach to high-level waste management at Hanford was a combination of waste fractionalization and in-tank solidification. Although continued tank storage was cheaper, the

higher cost of fractionalization was considered justified by the increased certainty of long-term confinement of the hazardous nuclides and by increased capability for fission product recovery. Research and development efforts were therefore being directed towards acquiring the necessary technology. Mr. Tomlinson, however, pointed out that these conclusions applied specifically to Hanford and were not necessarily applicable to other sites.

In a review of present and future programmes in the United States, W. G. Belter (USA) said: "While it appears that interim storage can be accomplished economically in tanks of present-day design at sites having suitable geologic and hydrologic conditions, the inherent safety restrictions of this operation such as potential leakage and the necessity of liquid waste transfer for periods of hundreds of years has resulted in a vigorous research and development programme in the US directed at engineering practical systems for ultimate disposal of these materials." The high-level waste management programme of the USAEC Reactor Development Division, said Mr. Belter, had followed two general approaches, namely, (1) conversion of these wastes to a solid oxide form or fixation of the activity in an inert, non-leachable solid carrier, and (2) the direct disposal of these wastes into specific geological formations, such as salt structures, impermeable crystalline rock, etc.

In a broad review of waste management practice and plans in the Soviet Union, B. Kolychev (USSR) said that at present the liquid wastes were first reduced in volume and their activity concentrated, the concentrated wastes being thereafter channelled into stainless steel tanks. The Soviet scientists, however, realized that steel tanks could not be used for ultimate storage, and were therefore engaged in extensive research work aimed at converting the waste to a more suitable form for permanent burial. The use of salt beds had been considered, but was not regarded as suitable for liquid wastes or particularly advantageous for solid wastes. Burial of liquid wastes in deep geological formations, although very tempting, was a very difficult procedure to study and there could never be any certainty of obtaining entirely reliable results from the testing of this procedure. "We consider", said Mr. Kolychev, "that the most promising procedure for the safe burial of radioactive wastes consists in solidification methods and, more particularly, in vitrification, because it is by such techniques that we shall be able in the future to bury radioactive waste permanently and to do so with confidence and under reliable control".

Methods of Solidification

Scientists from the United States presented several papers on different methods of solidifying liquid radioactive wastes by calcination, i. e. by strong heating. In a paper by W. R. Regan, L. P. Hatch and R. F. Demish, it was pointed out that the

calcination of high-level aqueous wastes with a high salt content, in order to convert the salts to stable solid form, has important advantages, such as major reduction in waste volume and general simplification of long-range storage problems. The authors described a type of calciner, now under development at the Brookhaven National Laboratory, which operates on the principle of a rotary ball kiln. Basically, the unit consists of a slowly rotating horizontal tube or kiln externally heated by means of electric resistance heaters. The liquid waste is introduced into the kiln by a distribution nozzle, and falls on a hot, shallow bed of metal balls, and the calcined solids overflow from the discharge end of the kiln into a powder receiver.

Four other American scientists - W.E. Winsche, M.W. Davis, Jr., C.B. Goodlett, Jr., and E.S. Occhipinti - described a novel calcination process based on certain special properties of sulphur, which is being developed at the Savannah River Laboratory. In this process the aqueous acidic wastes are reacted with molten sulphur at 150°C so that the water and the volatile acids are driven off and the chemical compounds present in the waste are calcined and/or chemically reduced. Further calcination and/or reduction is achieved by heating the resulting sulphur-waste slurry at 400° to 444°C to drive off the sulphuric acid and residual water. Among other things, the molten sulphur serves as a heat transfer medium in this process. After the high temperature treatment, the sulphur-waste slurry is cooled to 120° - 150°C and transferred as a liquid to the final containment system where it is allowed to solidify.

Three other calcination methods described at the conference are known as the pot calcination process, the fluidized bed process, and the radiant heat spray process. The pot calcination process, which is being developed at the Oak Ridge National Laboratory, involves the evaporation of liquid wastes in a pot which would then serve as the final container for storage; calcination takes place at temperatures of 700° - 900°C . The fluidized bed process involves the injection of waste solution through spray nozzles into a fluidized bed wherein the water evaporates, the nitric acid decomposes to water and nitrogen oxide, and the metallic nitrates decompose to the corresponding metallic oxides. The radiant heat process consists of feeding liquid wastes through a pneumatic nozzle into the top of a tower; the walls are heated to 850°C by passing low voltage current through the entire column, with water droplets passing down the tower through successive zones of evaporation, drying and calcination.

Despite the advantages of calcination over the present practice of tank storage of liquid wastes, calcination - as pointed out earlier - cannot immobilize the wastes permanently; the oxides formed by calcination are not non-leachable and the radioactive sub-

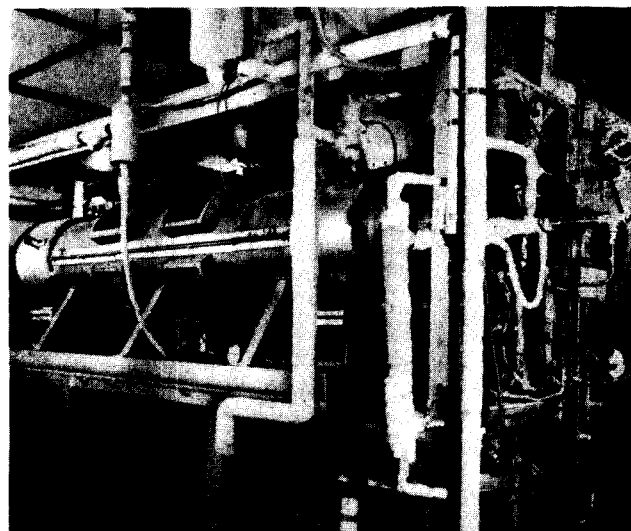
stances may therefore escape into the environment under certain circumstances. It was observed at the symposium that while calcination would represent a considerable advance on the present methods of storage, it could be no more than an intermediate step towards "ultimate" disposal; much more effective - in fact, virtually permanent - immobilization could be achieved by fixing the wastes in a suitable unleachable solid, such as glass. During the last few years, a great deal of research effort has been devoted to developing methods of fixing radioactive wastes in glass, and promising results were reported at the symposium by scientists from the United Kingdom, the United States and other countries.

It was pointed out that fixation of radioactive wastes in glass produces a final product with a number of desirable properties. For example, the glassy solid residues are non-volatile up to their formation temperatures, have good mechanical strength so that their containment is not completely dependent on the integrity of an outer container, have good thermal conductivity so that problems of heat transfer are minimized, and have a low solubility in environmental erosion media. The product need not be a true glass if it has these properties.

W.E. Clark and H.W. Godbee (USA) described current laboratory development work at Oak Ridge aimed at incorporating high-level wastes in high density insoluble glasses containing all the radioactive constituents in the solid. They said that experiments conducted so far indicated the technical feasibility of such a process.

A process of ceramic sponge fixation was described in the paper by W.G. Belter, referred to earlier. The process involves the preparation of a

A pilot plant scale rotary calciner for the solidification of high-level radioactive wastes. (Photo from paper by W.H. Regan, L.P. Hatch and R.F. Demish)





Photograph of a glass product containing radioactive waste. (From paper by M.N. Elliot, R. Gayler, J.R. Grover and W.H. Hardwick)

highly porous clay body which is fired to approximately 1100°C . This body is then soaked in the liquid waste, dried and re-soaked several times. It is finally fired at 1300°C to fix the radionuclides permanently in the ceramic material.

A paper by L. P. Hatch, G. C. Veth and E. J. Tuthill, also from the United States, reported on a process, now being developed at the Brookhaven National Laboratory, in which the entire conversion from the raw aqueous wastes to the final glass product would be carried out in an all-liquid system.

Reporting on pilot plant experience at Harwell in the fixation of radioactive waste in glass, M. N. Elliot, R. Gayler, J. R. Grover and W. H. Hardwick (UK) gave details of a process which aimed at converting highly active fission product wastes to a leach-resistant glass in stainless steel cylinders incorporating up to 40 per cent of waste oxides. The cylinders, they said, formed the basis of a storage system capable of lasting for centuries. Pilot plant operation had demonstrated the practicability of the process and many successful runs had been completed, producing 50 kg blocks of leach-resistant glass from low activity simulated waste solution. The next step was to operate with solutions that were sufficiently radioactive to enable decontamination factors to be measured for the important fission products and to prove the techniques of remote operation. A pilot plant to produce glass blocks containing 1000 curies had been designed and built.

A paper on the incorporation of radioisotopes into melted silicates, presented by J. Rálková and J. Saidl (Czechoslovakia), gave an account of research

work aimed at selecting melted rock and glass-type silicate materials suitable for the incorporation of biologically dangerous and long-lived radioisotopes, and determining the factors that could influence the rate of release of the incorporated isotopes.

Other Topics

Where suitable geological formations are available, it may be possible to store high-level wastes without any treatment, the characteristics of the storage places providing an adequate shield against the escape of the wastes into the environment. In the United States a number of studies have been made on the possible disposal of both liquid and solid high-activity wastes in salt structures. Natural salt formations have several advantages as repositories for radioactive wastes. Among these are the impermeability of salt due to its plastic properties, good thermal conductivity, good structural strength and "its non-association with usable ground water resources". American scientists reported that the storage of high-level wastes in salt mines appeared attractive, although certain aspects of the procedure required further investigation. For the disposal of solid wastes, one main uncertainty is the combined effect of heat and pressure on the structural stability of mined openings. As for the disposal of liquid wastes, certain problems arise from the radiolytic production of gases and alterations in the salt mine cavities.

Among other subjects discussed at the symposium was the shipment of large quantities of useful radionuclides extracted from the wastes. Some of the radioactive fission products, such as strontium-90, caesium-137, and cerium-144, have important uses as both heat and radiation sources. Because of the remote locations of the sources of supply of these fission products, it is necessary to package and ship bulk lots of these nuclides. Shipments of this type are subject to several types of potential accidents, and some of the problems that may arise in the course of such shipment were examined at the symposium.

Another important subject that attracted some attention during the discussions was the cost of waste management procedures. An economic evaluation of tank storage, pot calcination and shipping of radioactive wastes from the reprocessing of power reactor fuels was presented by J. J. Perona, R. L. Bradshaw, J. T. Roberts and J. O. Blomeke (USA).

Commenting on the cost factors of waste management, W. J. Belter in his review paper said: "The fraction of nuclear power costs allowable for waste disposal has not been established, and no realistic cost estimate for the ultimate disposal methods is available at this time. However, the cost of storing wastes in tanks of present design on a 'perpetual care' basis has been estimated to lie

between 0.1 and 0.15 mill/kWh of nuclear electricity produced. This cost approximates only one to two per cent of the cost of nuclear power in an 8 - 10 mill/kWh economy. The ORNL engineering and economic studies indicate that the total cost of interim liquid storage, pot calcination, and shipping over a 3000 mile round trip can be as low as 0.02 mill/kWh(e). Tentative costs for a large-scale

disposal operation for calcined solids in salt deposits are estimated at 0.01 mill/kWh(e). Based on past laboratory and engineering scale cold unit operation data, and on an expected successful field demonstration and testing programme with high activity wastes, it is firmly believed that waste management operations should not constitute a major obstacle to the development of economical nuclear power."

MEDICAL USES OF RADIOACTIVE CALCIUM

REVIEW OF AN IAEA PROGRAMME TO PROMOTE THE APPLICATIONS OF CALCIUM-47

Calcium plays a number of biologically essential roles, which have long been under investigation by various techniques available to medical science. One of the most important of these techniques is radioactive tracer analysis, i.e. study of the functions of calcium within the body with the help of a radioactive isotope of the element.

The calcium-47 programme of the International Atomic Energy Agency is intended to promote these investigations by facilitating the production and use of this isotope. The importance attached to calcium-47 is due to the special properties of this isotope, which make it the most valuable tool for many calcium studies by the radioactive tracer method.

Calcium as found in nature is a stable element and has six isotopes. The most common of these six stable isotopes is calcium-40, which constitutes about 97 per cent of natural calcium. The other stable isotopes are rare, and calcium-46, a stable isotope which is of particular interest in the production of the radioactive isotope calcium-47, has an abundance of only 0.003 per cent.

Advantages of Ca⁴⁷ and Methods of Production

Apart from the stable isotopes, at least six radioactive isotopes of calcium have been artificially produced. Three of these have properties which make them of possible use as radioactive tracers. Calcium-45 has been the most prominent of these three, and has been used extensively during the last 15 years. Calcium-49 has also been employed. But for many purposes the most suitable is calcium-47. Why this is so is explained by a comparison of the physical characteristics of these three isotopes.

Calcium-45 has a half-life of about 160 days and emits low energy beta radiation which is completely

absorbed in a layer of matter less than 1 mm thick. The relatively long half-life of the isotope results, in most human studies, in an undesirably prolonged irradiation of the subject and the isotope is therefore considered rather hazardous for internal administration. Also, the low energy level of its radiation often makes measurements difficult. Calcium-49, on the other hand, has a half-life of only nine minutes, which is too short for biological tracer applications, although convenient for certain other purposes. Calcium-47 has a half-life of about five days, which is long enough for numerous biological investigations and short enough to ensure that the person subjected to the investigation is not under prolonged irradiation. It emits gamma rays in addition to beta rays, and since the gamma rays are capable of penetrating many centimeters of tissue, determination of calcium-47 in the body is possible by external measurements. Furthermore, measurement of the calcium-47 content even of isolated samples, such as a few milliliters of blood, is far more convenient than that of calcium-45. Thus calcium-47 has distinctive properties which allow its use in biological investigations where calcium-45 would be unsuitable, and some of these uses are of considerable medical significance.

Unfortunately, calcium-47 is difficult to produce. The irradiation of ordinary calcium in a nuclear reactor, of course, results in the capture of some neutrons by the stable isotope calcium-46, which thereby is converted to calcium-47. However, because calcium-46 is extremely rare, the amount of calcium-47 produced in this way is insignificant compared with the amount of calcium-45, which is produced from calcium-44, a more abundant stable isotope. There are several other reactions by which calcium-47 can be produced, but they are all very expensive. At present the most economic approach is a sequence of two steps. First, calcium-