EXPERIENCE WITH INCINERATION OF LOW RADIOACTIVE WASTE IN EUROPE

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INTRODUCTION

The operation of nuclear power plants and nuclear research centers as well as the application of radioactive isotopes in the industry and in research laboratories, produce, a high volume of low radioactive waste. The conditioning, transportation, storage and final disposal of the different types of waste depends mainly on safety requirements and economic considerations.

In Europe a high volume reduction of the low radioactive waste has been essential since the beginning of nuclear technology activities, because the dense population, climate conditions and a high groundwater table in most areas exclude shallow land burial.

The United States has had success with shallow land burial of low radioactive solid waste. However, because of recent restrictions and problems with some burial sites, this country must also consider new technologies.

The composition of the waste depends on the sources. But as a rule of thumb, it can be assumed that about two-thirds or more of the solid low radioactive waste can be considered as combustible waste consisting of paper, clothings, plastics, rubber, and in some cases, organic materials like carcasses and dung.

The incineration of combustible waste is a very effective technique for reducing the volume. The volume reduction factor ranges from 20 to more than 100 depending on the amount of non-combustible material in the waste. A high-volume reduction factor can be achieved by sorting the waste at the collecting site or, as an additional step, before incineration.

The incineration of combustible waste has been favored in Europe for many years. Different types of incinerators have been developed in the Federal Republic of Germany, Belgium, the United Kingdom and France. Switzerland, Austria and Japan bought German incinerators.

DESIGN ASPECTS

For the incineration of combustible domestic and industrial waste, several types of incinerators with very large throughput (up to about 100 t/h) and an extremely simple flue-gas clean-up system are in operation. For the combustion of radioactive waste, however, much smaller units with a throughput between 50 and about 400 kg/h are necessary. The off-gas cleaning system must be very effective.

In contrast to common incinerators, the radioactive units should be operated at a small negative pressure to ensure that the radioactivity cannot leak out of the furnace.

Finally, the design should be as simple as possible to ensure easy maintenance and repair, while taking into consideration the possibility of radioactive contamination.

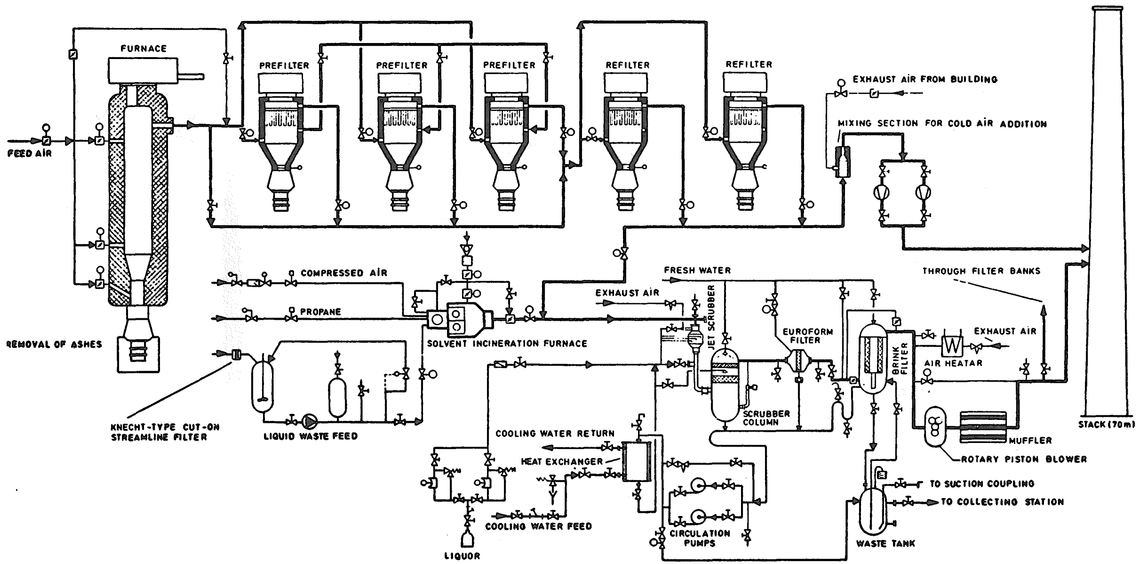
Due to these reasons it is advisable to develop tailor-made systems for the incineration of radioactive waste.

THE KARLSRUHE SHAFT FURNACE

The Karlsruhe Incineration Plant[1] consists of a very simple shaft furnace and a relatively expansive off-gas cleaning system. Its capacity is about 60 kg/h. Since February, 1971, the plant has been run for more than

Fig. 1

Incineration plant for radioactive waste
state as of September 1977
Kernforschungszentrum Karlsruhe
Federal Repuplic of Germany
Decontamination Department



5.10.1977
Kaufmann

20,000 h with an availability of about 81 %. More than 1000 t of solid and 150 m^3 of organic liquid waste were burnt.

The flowsheet of the plant is shown in Fig. 1. The waste is burned in a cylindrical shaft furnace with an inner diameter of 1 m and a height of 6 m. The furnace is lined with 0.5 m thick refractory brick. In the bottom part this brick lining tapers off conically, ending in a small cylindrical chamber with a diameter of 400 mm. The bottom is closed by a damper. The combustion air is introduced at 3 points: above the damper, above the cone, and at the top of the furnace opposite the flue gas outlet.

The furnace is loaded from the top via a slide lock. The air supply is controlled manually in the two lower openings, but automatically in the upper opening as a function of the temperature at the furnace outlet. The ash is withdrawn from the bottom of the furnace once a day via the damper.

The off-gas cleaning system consists of several ceramic high-temperature filters, which are arranged parallel and in series, and a scrubber. The off-gas reaches one of three ceramic lined prefilters, each containing 91 silicon carbide filter elements. The off-gas passes through the filter elements from the outside to the inside at an operating temperature of 900°C. Unburnt particles are retained at the surface and can be burned completely. The resulting ash falls off the filters into the bottom part of the filter housing. Downstream from the prefilters the off-gas passes through so-called "refilters", which are very similar to the prefilters. The operating temperature is about 600°C. The main function is to increase filter efficiency and to provide a safety factor in case of defects in the prefilters. The filtered air is cooled down to about 250°C by addition of fresh air. For several years, the off-gas was discharged through the stack. In 1977, however, the plant was backfitted with a liquid scrubber.

The release rate of radioactivity was on the order of to 10^{-4} for α-active nuclides and 10^{-2} to about 10^{-3} for ß-activity. In the latter case especially, Cs-137 and Ru-106 are responsible for the low decontamination factor. After installation of the scrubber, the release rate seems to be reduced by a factor of 500.

The lifetime of the ceramic prefilter elements is between 300 and 600 hours, that of the so-called refilters, which are working at a lower temperature, between 1000 and 1500 hours. The valve cones and valve seats in the off-gas lines must be replaced after 2 or 3 years of operation.

Abroad, several incinerators of this design were built, are being built, or will be built by several companies under licence agreement. In Würenlingen, Switzerland, a 25 kg/h plant has been in operation since 1975. A nearly identical plant is under construction in Seibersdorf, Austria. One 50 kg/h plant and one 100 kg/h plant have been built in Tsuruga and in Tokai, Japan. More plants are to be built in the coming years.

THE SWISS INCINERATOR

In the Swiss Nuclear Research Centre EIR in Würenlingen, a 25 kg/h incineration plant has been built[2,3] according to the Karlsruhe design. PVC is excluded from the incineration, herefore, no scrubber has been installed. The filter system consits of two prefilters and one "refilter".

The plant began operating in October 1975. Since this time about 100 tons of waste have been burnt during 4 phases of operation. The volume reduction factor was about 65. In the first phase, some filter elements broke down after a relatively short time. After exchanging all the elements and gaining experience, no problems existed with the filter elements.

Some difficulties arose from the slag formation, which was favored by a high polyethylene content in the waste. However, the problems could be reduced by charging smaller batches of wastes and complete burnout of these batches.

The release rate from the plant was determined in several special experiments. It amounts to $5 \cdot 10^{-4}$ to

$7 \cdot 10^{-4}$ for Cs, $2 \cdot 10^{-3}$ for Co and $5 \cdot 10^{-3}$ to $6 \cdot 10^{-5}$ for Pu These results are surprising and cannot be explained completely.

The aerosol concentration in the discharged off-gas is on the order of 1 to 10 mg/m^3 STP, on the average about 5 mg/m^3 STP. This corresponds to about 100 g solids per hour.

THE JUELICH INCINERATION PROCESS

The Nuclear Research Centre Juelich has developed the Juelich Incineration Process for low active combustible waste[4]. The pilot plant has a capacity of about 100 kg/h. The plant is very compact. If the PVC-content of the waste can be limited to about 5% by weight, a dry off-gas cleaning system can be applied. The plant is very flexible with respect to the waste composition. Waste with an average heat of combustion of more than about 4,200 kJ/kg can be burned without additional fuel. The quality of the off-gas and of the ash is very good. Incombustible material such as glass bottles, spray cans, inorganic filter material, or metal mountings do not create any problem.

The incinerator consists of a pyrolysis chamber and an oxidation chamber, arranged one beneath the other and seperated by movable paddles (Fig. 2). The pyrolysis chamber is filled with waste via a lock. According to the "burn-out" at the bottom, the waste passes down slowly and fresh waste is fed in. During the operation of the plant, horizontal and vertical gradients were established in the waste column.
The waste is gradually heated up to about 800°C. Step by step it is dried, decomposed, and partially gasified in reducing atmosphere.

The energy needed for the endotherm pyrolysis reaction results partly from hot off-gases and partly from the reaction with understoichiometric amounts of primary air which is fed into the pyrolysis chamber. The pyrolysis products are sucked into the oxidation chamber. While passing the hottest zone near the paddles, the high molecular compounds like tar and oils are cracked into lower molecular aliphatics and aromatics. In the oxidation chamber the pyrolysis gases are mixed with excessive air which is fed through the paddles, and burned completely. Also ash and nongasified pyrolysis coke falls through the gaps between the paddles and the wall of the incinerator into the oxidation chamber, where it can burn out completely in an oxidizing atmosphere. Through a lock at the bottom of the oxidation chamber, the ash is withdrawn chargewise into a drum. The hot off-gas passes through a high temperature filter mat, is cooled down to about 250°C in a heat exchanger, passes through a steel wool filter, is mixed with fresh air down to a temperature of 120°C, and is finally purified by HEPA filters.

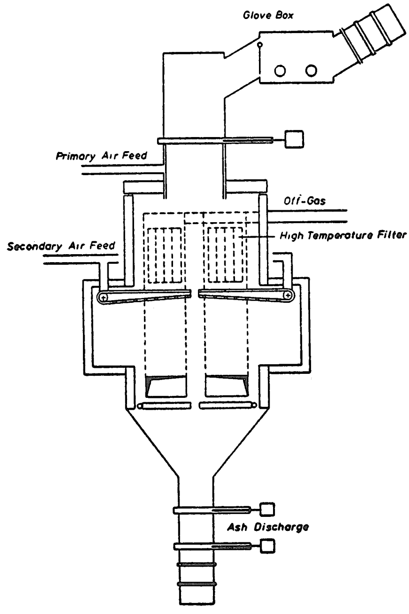


Fig.: 2 Juelich Incinerator[4]

The incinerator can be operated and controlled very easily. Because the waste is gradually heated and decomposed, large changes in the composition of the waste are equalized. A large packet of polyethylene with a very high heat of combustion is decomposed during several hours, so that the temperature in the pyrolysis chamber is not increased abnormally. This behavior is called "thermal homogenization". Therefore, a mechanical homogenization, e.g. with a shredder, is unnecessary.

In spite of the very slow reaction, the temperature profil in the pyrolysis chamber can be controlled very easily by controlled addition of the primary air, which reacts with the pyrolysis gases. Waste with an average heat of combustion of more than 4,200 kJ/kg (=1,000 kcal/kg) can be burned without additional heating. Two gas or oil burners are used only during the start-up phase.

Since 1977 the pilot plant was 150 days or about 1500 hours in operation. During this time, about 82 t of low radioactive waste containing 25.7 t mixed waste, 14.1 t wood, 25.8 t paper, 2.3 t clinical waste, 15.4 t carcasses and smaller amounts of plastics, graphite and tires were burnt. During the total operation no serious problems arose.

After about 600 hours, the brick liner was changed to improve the heat transfer from the flue gas to the pyrolysis chamber. With the exception of some minor cracks, which would have been repaired easily, no damage to the liner could be found.

The lifetime of the high-temperature filter mats has been estimated to about 1,000 hours. The lifetime of the steel wool filter, which acts as a safety prefilter for the HEPA filters, is about 250 hours; that of the HEPA filters more than 100 hours.

The off-gas contains less than 100 ppm CO and less than 50 ppm organic compounds. These values may be exceeded only during the start-up phase.

The ash is completely burnt out and sterile. The radioactive release is very low. Special experiments were made with known activities of 137-Cs, 85-Sr and 152-Eu. The release factor was $9\cdot10^{-7}$ for Cs and less than $5\cdot10^{-7}$ for Eu and Sr.

Based on the good results with the Juelich installation, more plants have been put into operation or are under construction for the incineration of hospital and special industrial (plastics, rubber) wastes.

THE FLK HIGH TEMPERATURE SLAGGING INCINERATOR

The FLK process [5,6,7,8] has been developed in Germany for the incineration of domestic and industrial waste. At Hino, Toyota, in Japan an incinerator began operating in 1975.

In the same year, a 100 kg/h plant for radioactive waste was installed at Mol, Belgium. The unit has been in active operation since 1978. The composition of the waste is about 30% paper, 15% plastics (PE), 10% ion exchange resins, 10% metals, 6% sand, 2% glass, 17% water and 10% sludge. The minimum content of incombustible slag forming material is about 20% by weight. Otherwise, no continuous melting layer at the bottom of the flame chamber can be achieved. Therefore, the FLK process should be used if larger amounts of sludges must be burned.

The FLK furnace operates at 1400 to 1600^{o}C with a small surplus of air. The molten residue drops into a water bath forming granules with a very low solubility. The main components of the FLK furnace are shown schematically in Fig. 3.

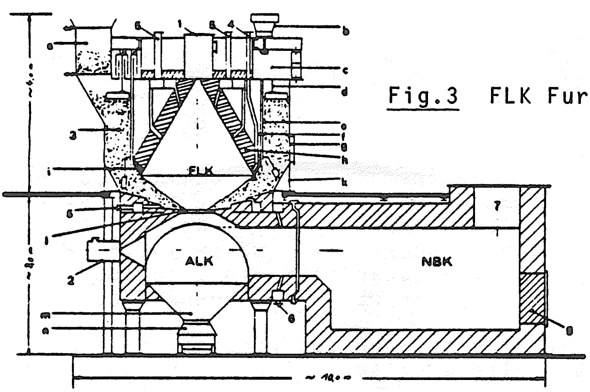


Fig.3 FLK Furnace[7]

The furnace consists of a flame chamber (FLK), an outlet chamber (ALK) and an afterburner chamber (NBK).

The waste is fed into an annular gap (3) between an outer fixed shell and an inner rotating cylinder which is equipped with drivers for the distribution and the transport of the waste. At the bottom of the flame chamber, the waste is heated by a burner (1) mounted concentrically at the top of the flame chamber. The pyrolysis gases produced by the degradation of the waste are burned by addition of preheated air (5). At temperatures between 1400 and 1600°C, the incombustible residue melts and drops into the outlet chamber (ALK), which is filled with water in the lower part of the chamber. The off-gas is mixed with secondary air (6) and is completely oxidized in the afterburner chamber (NBK) at about 900°C. In contrast to other incinerators for radioactive waste, the FLK furnace operates at a positve pressure of about 200 to 500 Pa (20 to 50 mm of water column).

The dust concentration in the off-gas is on the order of 80 to 300 mg/m^3 STP. This is much lower than expected, presumably because the molten slag acts as a catcher for the particles.

Also the volatilization of heavy metals was lower than suspected. Japanese measurements have shown that during the incineration of sludges, 87.2% Cd, 31.0% Pb, 88.2% and 92.4% Cu were retained in the granules. The off-gas coming from the afterburner chamber is cooled down to 700°C, filtered by a sand filter, cooled down to 300°C and scrubbed with an alkaline solution. Finally it is cleaned by HEPA filters.

BRITISH INCINERATORS

Harwell started incinerating low-activity solids in 1950. Since that date several incinerators have been in operation[10]. At the present time, Wellman Incandescent Ltd. offers two standard sizes of an incinerator for reactor wastes with capacities of up to 150 lb/h and up to 50 lb/h. One incinerator has been in operation since 1977[6,9].

The Wellman incinerator consists of a main primary chamber with a grate, below which is a secondary chamber and second grate to prevent unburned material from dropping into the ash pit. The ash residues are removed by a screw conveyor. The waste is burned with an excess of air. The off-gas coming from the incinerator chamber is oxidized completely in an afterburner chamber after addition of secondary air. In the existing plant, the off-gas is cooled by a heat exchanger. Because of extensive corrosion, more recent designs provide instead a direct water quench as part of the wet gas scrubber. The scrubber solution is maintained at a pH between 6 and 8 thus minimizing corrosion and allowing a re-circulation of the solution. The scrubbed off-gas passes through HEPA filters and is discharged into the atmosphere. No details are known at the present time.

INCINERATOR AT MARCOULE, FRANCE

The Marcoule Incinerator is designed for the combustion of low radioactive waste containing about 56% rags, 16% wood, 13% polyethylene and PVC, 10% paper, and 5% rubber. The capacity is 80 kg/h. Also oils can be burned [10,11,12].

The furnace (Fig. 4) consists of a combustion chamber (1.7 m^3), an afterburner chamber (0.8 m^3) and an expansion chamber (0.7 m^3). Four propane burners maintain the combustion chamber at 1150°C and the after-burner chamber at 1100°C. The off-gas is cooled by heat exchangers in two steps to 400 and 150°C. Then the gas is routed via a cyclone, a bag filter, prefilters, and HEPA filters to a neutralizing tower.

The ashes are withdrawn with water via a channel (wet method) or using a glove box fitted with a rake (dry method). 92% of the radioactivity is retained in the ash, 5% is found in the dust and 3% in the water pool. The main activity in the dust is Ru-Rh (41%) and sulfur (24%). The water contained 92% S-35 and 8% Ru-Rh.

From the end of 1963 to 1968, about 500 tons were burnt after sorting. The furnace operated 6 days a week. The activity of the waste ranged from 9 to 15 mCi/ ton. The rest activity in the purified off-gas was lower than the environmental activity concentration.

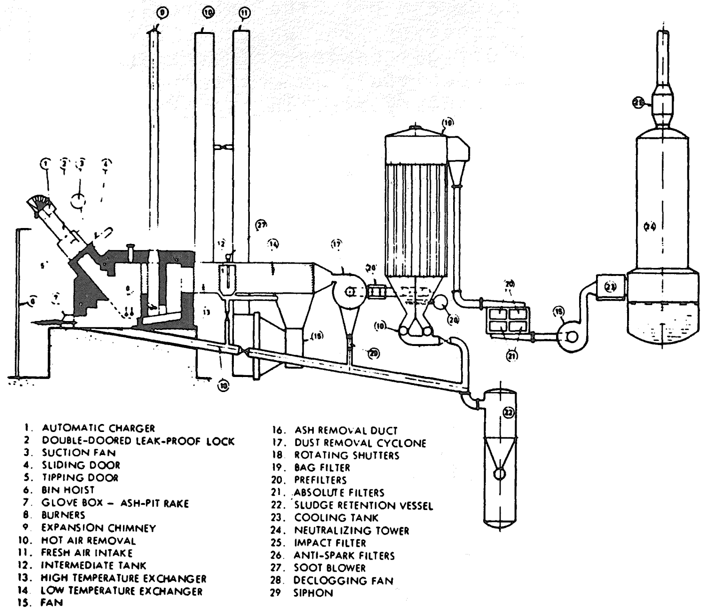


Fig.4 General layout of incinerator plant, Marcoule.[10]

If oils and solvents are burned together with the solid waste the propane consumption can be reduced remarkably.

A high PVC content in the waste causes severe corrosion. Therefore, PVC was replaced by polyethylene as much as possible. This solution to the problem seemed to be more favorable than the use of high corrosion resistant materials.

THE C.E.C. INCINERATOR FOR SPECIAL WASTES

SociétéCarbonisation, Entreprise et Cēramique built incinerators in Strasbourg, France, and in Japan[13]. The capacity ranges from 20 to 120 kg/h. The incinerator consists of a charging shaft, a combustion chamber and an afterburner chamber.

At the bottom of the charging shaft there is a sloping grate which transports the wastes into the combustion chamber. Primary air is fed under the grate. The ash falls down through the grate into the ash pit. The combustion chamber is equipped with oil or gas burners for complete oxidation. The off-gas purification system contains a high-temperature filter with metal filter elements coated with asbestos filter material and HEPA filters.

The Strasbourg Plant is designed for the incineration of carcasses containing mainly C-14 and tritium. Therefore the results are not specific for the incineration of "normal" solid wastes from nuclear technology and scientific laboratories. As for the experience with the Japanese plants, no data are known.

CONCLUSIONS

Nearly 20 years experience with the technique of incinerating low radioactive solid waste has shown that it is a favorable way to reduce the volume of the waste. However some problems remain. A high PVC content causes severe corrosion in all parts, especially in the off-gas lines and heat exchanges. It is more advantageous to reduce the PVC content than to use high corrosion resistant material. The reduction can be achieved by replacing PVC with PE as much as possible (France), by administrative means (Juelich/Germany) or by sorting (Wuerenlingen/Switzerland).

A good burn-out of the off-gas is essential. Soot and tar should be reduced to a minimum, because in some cases soot and tar deposits caused a fire "at the wrong place" in the incinerator. Means for the prevention of such incidents are, for instance, high-temperature ceramic filter elements (Karlsruhe), high temperature filter mats(Juelich) which retain unburded material until complete combustion, extensive afterburner chambers with additional gas burners (Marcoule), or steel wool filters as spark catchers (Juelich). Most of these means, however, fail if the waste is fed into the hot zone of the incinerator and a very large volume of pyrolysis gas is formed. Then the amount of combustion air is insufficient for a short time and high molecular pyrolysis compounds leave the incinerator. This is the case with many incinerators. In the Juelich Incineration Process, however, the waste is fed onto the cold waste column and is heated gradually. The pyrolysis products are sucked throught the hottest zone of the furnace and are cracked to low molecular compounds. Therefore, no porblems exist with tar and soot and a very simple and compact off-gas purification system can be applied. Our experience has proven that excellent combustion and a high quality off-gas is better and cheaper than an extended and expansive off-gas purification system.

A goodburn-out of the gases combined with low PVC content in the waste facilitates the off-gas purification the most, because in this case a completely dry filter system can be applied thereby avoiding the generation of a secondary liquid waste stream.

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