

OPERATIONAL EXPERIENCE ON BITUMINIZATION IN KOREA

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ABSTRACT

Bituminization process using the thin film evaporator has been in operation at Korea Atomic Energy Research Institute (KAERI) since 1991 through test-operation between 1987-1990. The evaporation capacity of the thin film evaporator is 40 L/hr and flow rate of bituminized products is about 30 L/hr under normal condition. Bituminization is carried out using a straight bitumen (AC 40-50) at evaporator temperature of 200-220°C.

In this paper, bituminization process installed at KAERI and operational experience obtained during test-operation are described and an effective treatment method of liquid waste and spent resins is presented.

INTRODUCTION

The Korea Atomic Energy Research Institute built the Radwaste Treatment Facility (RWTF) in 1987 with design of SGN in France. It includes processes of bituminization, liquid radwastes concentration, and solid radwastes treatment. However most of processes in RWTF has not been normally operated until 1990 because treatment method for very low liquid radwaste (VLAW) generated from them was not chosen. Solar evaporation was adopted to process VLAW, and the facility was constructed from 1988 to 1989, and was successfully operated in 1990. It led to normal operation of the RWTF, at last, operation of the RWTF was licensed by the Korea Institute of Nuclear Safety (KINS) on March, 1991.

Bituminization process using the thin film evaporator, the key part of the RWTF, was tested by using simulated wastes between 1987-1990, in order to know whether it can guarantee the normal operation of the RWTF. It could be operated using low level liquid radwastes in the beginning of 1991. After normal operation, 10 drums of bituminized waste form were produced in 1991. However, in spite of a well-known and proven technology, bituminization process using a thin film evaporator was faced with several difficulties during test-operation. Those difficulties are described and discussed.

BITUMINIZATION PROCESS

A flow diagram of bituminization process installed in the RWTF is shown in Fig. 1. This process is very similar to bituminization process at Barsebek power plant in Sweden

(1), except differences in treatment capacity and some apparatus. The process consists of 6 major unit processes.

Wastes Storage and Feeding System

Liquid waste is directly transported into and stored in a 5 m³ feeding tank, equipped with a 2-speed stirrer and a steam-heating coil. Slurry of spent resins with demineralized water is transported into a 3 m³ resin storage tank. Powdered resins are transported into a feeding tank prior to be fed into the thin film evaporator, and bead resins are also poured into the same tank after grinding using a the wet grinder. Liquid waste, or resin slurry are heated up to 60°C in the feeding tank and fed into the thin film evaporator with a constant rate of 0-40 L/hr by a PAAC pump and a metering wheel. An emulsifying agent is added during feeding of wastes, if necessary.

Bitumen Feeding System

Bitumen is melted at 240°C and stored at 140°C in a 600 L reservoir. Molten bitumen is supplied with the constant rate of 6-24 L/hr into the thin film evaporator through the heat traced pipe by the geared pump.

Heating Oil Supply System

Most of heat required to operate bituminization process is supplied by heated oil. The electric heater with the capacity of 90 kW generates energy to heat up the oil. There are 2 loops in the heating oil circuit; One loop is to store and transport the molten bitumen with keeping the temperature at 140°C, and the second one is to operate the evaporator and melt the bitumen at 240°C.

Thin Film Evaporator

LUWA NL4-150 type thin film evaporator is installed in the RWTF and its evaporation capacity is 40 L/hr at 240°C. Liquid waste, or spent resins slurry to be solidified and hot bitumen are simultaneously introduced into the distribution ring which is the upper part of the thin film evaporator. Mixture of wastes and bitumen is projected with high turbulence by the distribution ring and the rotor on the heating surface in the form of a regular film. During the spread mixture goes down by gravity, evaporation occurs strongly and dehydrated wastes are homogeneously mixed with bitumen. After 1 or 2 minutes, molten product reaches the lower part of the evaporator heated up to 160°C, and flows from out the

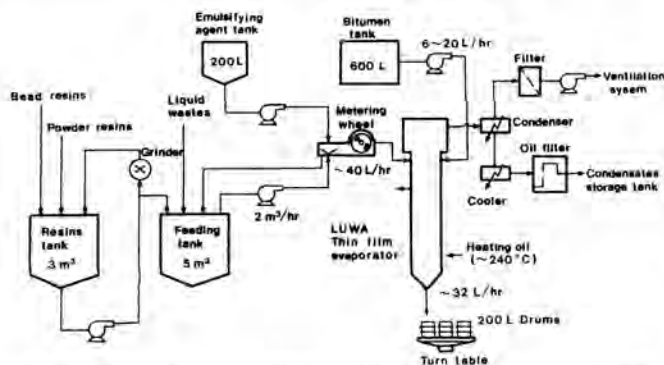


Fig. 1. Flow diagram of bituminization process at RWTF.

bottom of the evaporator into 200 L drum located under the evaporator.

Condensation and Discharge System

The steam generated from the evaporator flows counter-currently upward and out. The steam is condensed in the condenser and the condensate is passed through a cooler and a cartridge filter and storage at the low level liquid radwastes storage tank in the RWTF. Non condensible gases in the condenser are discharged into ventilation system equipped with HEPA filters.

Drumming System

The drums, which are stood on the 9 position turntable, are filled in two steps. At the first step, a drum is filled about 60% and replaced next drum. At the second step, that drum is returned to the filling position after about 36 hrs and then completely filled. The product is cooled down again for about 27 hrs, and crimped and removed.

OPERATIONAL EXPERIENCE

Bitumen Compatibility

Two kinds of straight bitumen (AC 40-50 and AC 85-100) and three kinds of blown bitumen (40-50 grade, 50-60 grade, and 60-70 grade in penetration value, 1/10 mm) were used with simulated wastes to confirm operational compatibility in bituminization using the thin film evaporator. SGN recommended to use the Mexphalt 40/50 graded straight bitumen, however blown bitumen was tested because Mexphalt 40/50 graded straight bitumen was not produced at that time in Korea. Fig. 2 shows the results during the operation. Fig. 2-a shows flow pattern of the molten bitumen. As soon as simulated waste is fed into the thin film evaporator, the amount of flow rate remarkably decreased (Fig. 2-b). After a few minutes, the flow was stopped (Fig. 2-c) with some noise and vibration of rotor, and then inhomogeneous mixture of waste without was not dehydration and bitumen were dropped out as a form of lumps (Fig. 2-d). At the same time, a large amount of steam is generated from the bottom of the evaporator. Therefore, it was impossible to operate normally and to produce the homogeneous waste forms. The cause could not be manifested yet, however, is expected to be the difference of viscosity between straight bitumen and brown bitumen, or the other properties such as foaming of blown bitumen by contacting with water, or mechanical faults of the evaporator. In spite of much efforts such as variation of evaporator temperature, rotor speed, and addition of surfactant, the problem was not resolved.

Test operation was retried by using straight bitumens (AC 40-50 and AC 85-100). AC 40-50 graded bitumen could be

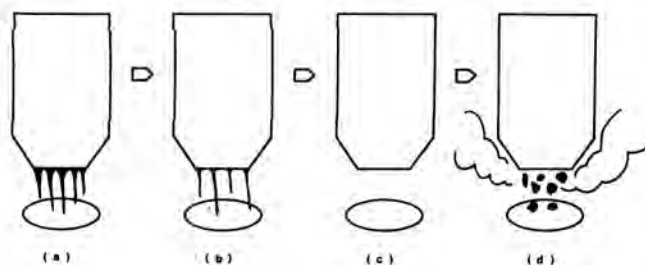


Fig. 2. Trouble in bituminization by using a blown bitumen.

obtained only from a manufacturer since it was not widely used in Korea. Bituminization was successfully carried out in these cases. The product was homogeneous and its moisture content was less than 0.5%.

Although some paper (2) reported that blown bitumen was used in bituminization process using the thin film evaporator, it was found, in this work, that blown bitumen was not compatible in bituminization using the thin film evaporator. AC 85-100 graded straight bitumen was suitable in operation, but it was too soft and form stability was doubtful. In conclusion, AC 40-50 graded straight bitumen should be used in bituminization process in spite of difficulty of purchase.

Operation Temperature

The heating temperature of the evaporator didn't remarkably affect the bituminization of liquid waste in the range of 200-240°C. In case of bituminization of spent resins, however, the viscosity of molten product from the bottom of the evaporator increased above 230°C and the normal flow was not formed as shown in Fig. 3. A number of small bubbles were also observed in the products. As operation was carried out after lowering the temperature into the range of 200-220°C, the normal flow could be maintained. Probably, degradation



(a)



(b)

Fig. 3. Bituminization of spent resins with straight bitumen (AC 40-50), a) at 240°C, b) at 220°C.

of resins occurs when evaporator temperature is higher than 230°C and it could be one of the reasons for abnormal flow.

Bituminization of Liquid Radwaste

Bituminization was carried out using liquid radwaste after completion of test operation. Results are listed in Table 1, which shows the operating condition, the characteristics of wastes and the final product. In spite of high volume reduction factor, solid content in a waste form (end product) is only about 17% due to very low content of liquid wastes. Therefore, it is considered that another concentration prior to bituminization will be required for effective operation. In this work, however, any concentration prior to bituminization was not performed since the total amount of liquid radwaste was not sufficient to be concentrated by another evaporator (in the RWTF). If salt content of liquid radwaste is 30%, it will be possible to produce the bituminized product which contains solid content of 40% at the rate of 30 L/hr.

TABLE I

Results of Bituminization for Liquid Waste

	Liquid Wastes	Bituminized Product
Concentration (or Content) of salts (NaNO ₃), %	3	17
NH ₄ concentration	< 0.1 g/L	-
NO ₂ concentration	< 2E-5 N	-
Free water	-	< 0.5 %
Radioactivity	2.8E-2 uCi/cc	0.05 Ci/drum
Surface Dose Rate, mR/hr	-	10-20
Volume Reduction Factor:	5.6	
Decontamination factor:	about 1000	
Bitumen used:	straight bitumen, AC40-50	
Temperature of Bituminization:	230°C	

A Consideration for Effective Operation

When bituminizing the liquid waste and spent resins, it is possible to bituminize them respectively or together after mixing if there is no problem in mixing. The effect of volume reduction, treatment time required in bituminization, final volumes of products are compared in both cases for effective operation.

If liquid radwaste and spent resins are bituminized, respectively, treatment time (Ts) required in bituminization, final volume of products (Ns) and average volume reduction factor (VRFs) are expressed as follows:

$$T_s = T_c + T_r = C / W_c + R / (a \cdot W_r \cdot X_r)$$

$$N_s = T_c \cdot P_c + T_r \cdot P_r = C \cdot (X_c / Y_c) + R / (a \cdot Y_r)$$

$$\begin{aligned} \text{VRFs} &= (C + R) / N_s \\ &= (C + R) / \{ C \cdot (X_c / Y_c) + R \cdot (1 / a \cdot Y_r) \} \\ &= (C + R) / (C / \text{VRF}_c + R / \text{VRF}_r) \end{aligned}$$

Where T_c: treatment time (hr) for bituminization of liquid waste, T_r: treatment time (hr) for bituminization of spent resin, C: volume of liquid waste (L), W_c: feed rate of liquid waste (0-40 L/hr), R: volume of wet spent resins (L), W_r: feed rate of resins-demineralized slurry (0-40 L/hr), a: volume ratio of wet resins and dry resins (about 2), X_r: fraction of dry resins in the slurry (less than 0.05), P_c: flow rate of products in bituminization of liquid waste (L/hr), P_r: flow rate of products in bituminization of spent resins (L/hr), X_c: fraction of salts in liquid waste, Y_c: fraction of salts in products, Y_r: fraction of dry resins in products, VRF_c: volume reduction factor for bituminization of liquid waste, VRF_r: volume reduction factor for bituminization of spent resins.

When liquid wastes and spent resins are mixed and bituminized, treatment time (T_m), final volume of product (N_m) and volume reduction factor (VRF_m) are also expressed as follows;

$$\text{For } R / (C + R) < \text{ or } = a \cdot X_r, \text{ max (about 0.1),}$$

$$T_m = (C + R) / W_m$$

$$N_m = T_m \cdot P_m = T_m \cdot [W_m \cdot \{ (X_c + X_r) / (Y_c + Y_r) \}]$$

$$\text{VRF}_m = (C + R) / N_m = (Y_c + Y_r) / (X_c + X_r)$$

$$\text{For } R / (C + R) > a \cdot X_r, \text{ max (about 0.1),}$$

$$T_m = (C + R') / W + (R - R') / (a \cdot W_r \cdot X_r)$$

$$N_m = (C + R') \cdot (X / Y) + (R - R') / (a \cdot Y_r)$$

$$\text{VRF}_m = (C + R) / N_m$$

$$= (C + R) / \{ (C + R) \cdot (X / Y) + (R - R') / (a \cdot Y_r) \}$$

Where W_m: feed rate of the mixture of liquid waste and spent resins (0-40 L/hr), X_c: fraction of salts in the mixture, X_r: fraction of dry spent resins in the mixture, X_{r,max}: Maximum permissible fraction of dry resins in the mixture (0.05), P_c: flow rate of products in bituminization of liquid waste (L/hr), P_m: flow rate of products in bituminization of the mixture of liquid waste and spent resins (L/hr), X: X_c + X_r, Y: Y_c + Y_r, R': (a · C · X_{r,max}) / (1 - a · X_{r,max}).

A result obtained using above equations, considering the operating condition (especially, such as capacities of feeding pumps), is shown in Fig 4. It indicates that the higher volume reduction factor can be obtained when liquid waste and spent resins are mixed and bituminized if salt content of liquid waste is below 10%, however, volume reduction factor is same in any cases if salt content is above 10%. A result of comparison with two cases about treatment time is shown in Fig. 5. It is always processed more rapidly when bituminizing after mixing liquid waste and spent resins. It means that bituminization after mixing liquid wastes and spent resins is more economical and the possibility of radiation exposure of operator can be also minimized. Furthermore, the stability of product increases since the content of spent resins in the product can maintained below than that of product when bituminizing only the resins.

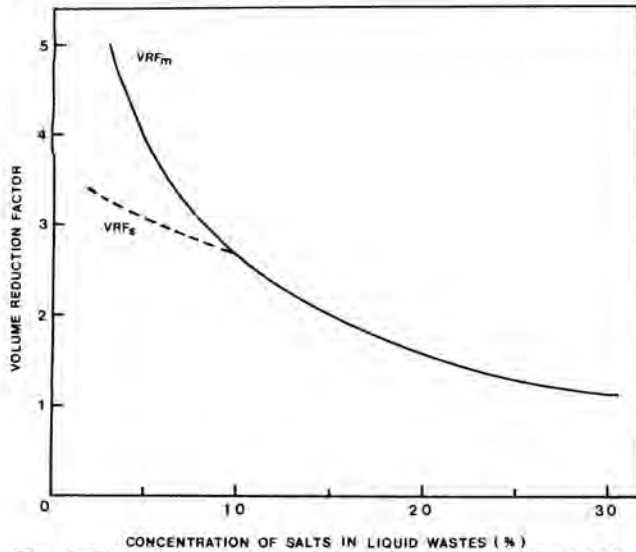


Fig. 4. Comparison with VRFs and VRFm (R/C = 0.11).

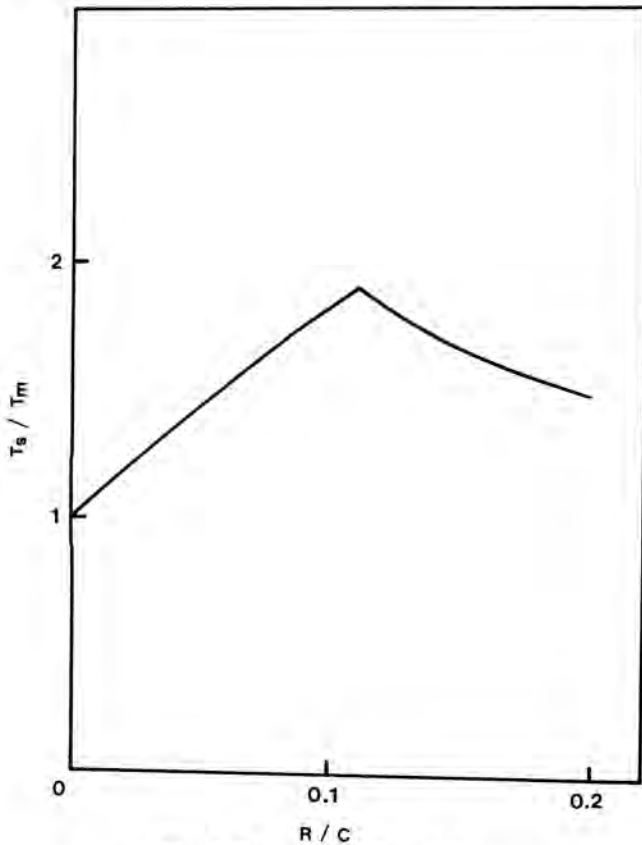


Fig. 5. Relation of Ts and Tm.

When liquid waste (as listed in Table I) and spent resins are bituminized after mixing, a result is listed in Table II. Limitation in solid content of product due to low salt content of liquid wastes can be overcome.

TABLE II

Results of Bituminization for Liquid Radwaste and Spent Resins after Mixing

	Bituminized product
Solids content	40%
Salts (NaNO ₃)	15%
Spent resins	25%
Radioactivity	0.06 Ci/drum
Surface dose rate	15-20
Volume reduction factor	5
Decontamination factor	1,500
Bitumen used	Straight Bitumen, AC40-50
Temperature of bitumenization	220°C

SUMMARY

At present, bituminization process is in operation by using AC 40-50 graded straight bitumen at 200-220°C. The use of blown bitumen is not considered since it gives abnormal operation. It is concluded that the process in which liquid waste and spent resins are bituminized after mixing is desirable in the view of volume reduction and treatment time. Studies on characteristics of bituminized products (such as leachability, mechanical strength, and biological property) are not performed yet, it will be soon started by research group at Facility for Waste Forms to be constructed in 1991.

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