

## A Study on the Bituminization Process of Radioactive Liquid Waste (II)

### The physical and chemical properties of bitumen-waste products

Sang Hoon Lee, Myung Hwan Yoon

Korea Atomic Energy Research Institute, Seoul, Korea

Moon Deuk Lee

College of Eng., Seoul National University, Seoul, Korea

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#### Abstract

The effects of temperature and pressure of leaching water on the leaching of radionuclides from bitumen-waste products were studied. The principal results are as follows: The fraction of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  leached for periods of up to 120 days at 8 atm was  $2.1 \times 10^{-6} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$  and  $6.02 \times 10^{-5} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$  respectively and at  $5^\circ\text{C}$ ,  $1.7 \times 10^{-5} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$  and  $4.01 \times 10^{-5} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$  respectively.

These values were lower than those in atmospheric pressure and room temperature.

No difference in the leaching rate with sea and distilled water was observed for the bitumen-waste products containing 40wt% salts. It appears that these results could be saved by improving safety in the dumping of sea. The effect of the softening point of pure asphalt or bitumen-waste product by  $^{60}\text{Co}$  irradiation was increased with increasing total dose. Irradiation of asphalts at a total dose of  $5.8 \times 10^8$  rad showed no evidence of volume and caused no swelling. The functional groups of blown asphalt by infrared spectra are also identified.

#### 요 약

중 준위 방사성 폐액의 증발 농축물을 불용성 물질로 고형화하기 위해 blown asphalt와 straight asphalt를 혼합사용하여 제조된 bitumen-waste 고화체로 부터 방사성핵종의 용출에 대한 고화체의 화학적 특성 및 용출 조건의 영향에 대해서 조사하였다.

고압 및 저온하에서 120일동안  $^{90}\text{Sr}$  및  $^{137}\text{Cs}$ 의 용출율은 상압 및 상온때 보다 낮은 용출율을 보였으며, 40wt%염을 함유한 고화체에 대한 인공 해수 및 증류수에 대한 용출율의 영향은 별로 차이점이 관찰되지 않았다.

순수 asphalt 및 고화체의 연화점은  $^{60}\text{Co}$  조사에 의해 총선량 증가와 함께 증가했으나,  $5.8 \times 10^6 \text{rad}$ , 까지 조사된 asphalt는 부피 증가나 팽윤 현상이 나타나지 않았다. 또한 Infrared spectra에 의해 blown asphalt의 functional group을 확인하였고, 계속 방사선에 의한 영향등을 조사중이다.

### 1. Introduction

The importance of radioactive waste treatment has been increased as the nuclear industry grows apace. Various kinds of experiments have actively been implemented on the economical treatment methods of radioactive waste for ultimate disposal in such a manner that the environment should not be contaminated by the fission products

The intermediate level wastes generated in nuclear power plant or nuclear fuel reprocessing plant generally have been stored in tanks or mixed with cement.

These wastes are characterized by their modest levels of radioactivity and by their high salt or solids content. From this point of view, studies have been carried out on solidification methods, and on the leachability of the solidified products of the radioactive concentrates from evaporators and chemical precipitators containing various radionuclides.

However, neither tank storage nor solidification in cement is completely satisfactory.

Tank storage is only a temporary measure, and the products formed by mixing the waste with cement are only moderately insoluble and represent a volume increase. In addition, most operation with cement are difficult to achieve good mixing and are cumbersome,

Accordingly, the availability of cementation process is only limited to the disposal of comparatively low level wastes. But, compared with the cementation process, a promising recent development is the use of asphalt or glass to solidify and insolubilize these wastes.

Though vitreous materials produced through the solidification of wastes are the most resistant to leaching of the activity by water, thus making it possible to incorporate the high-level radioactive waste into them, the vitrification process needs the high temperature more than  $1000^\circ\text{C}$  and costs greatly. Therefore, the technique of bituminizing radioactive wastes has come to be used on a wider scale in various countries over the last several years. Immobilization of wastes by asphalt incorporation before burial or ocean storage can reduce not only treatment costs but also the release of radioactivity to the environment. Plants for incorporation intermediate level waste in asphalt were already in operation at Mol Belgium, and Marcoule, France. Works on the development of bitumen-waste incorporation that has been carried out in various countries result in three different methods: 1) a fixing method for the nonemulsified asphalt-waste that has been developed by Eurochemie, Mol, Belgium,<sup>1, 2)</sup> 2) a method for temporary emulsion bituminization of radioactive sludge by the Marcoule Nuclear Research Center, France,<sup>3, 5)</sup> and 3) a me-

thod for the emulsified bitumen-waste incorporation by ORNL, USA.<sup>6, 8)</sup>

Asphalts into which the most part of wastes are incorporated are the two types of straight-run distilled asphalt (straight asphalt) and emulsified asphalt.

However, the previous report by KAERI<sup>11)</sup> used the blown asphalt whose softening point was higher than that of the straight asphalt, but this study used mixed asphalt (blown asphalt+straight asphalt) as a solidifying material.

In a study of the absorbance of asphaltic materials, J. E. Schmeyer<sup>9)</sup> in 1958 attempted to explain certain differences in the properties of asphaltic materials on the basis of the ultraviolet and infrared spectra, and the research described by Beitchman<sup>10)</sup> was the first successful effort to produce infrared spectra of asphalt films without resorting to solvents, mulls, pellets, or salt-crystal supports.

Accordingly, the special areas investigated in this report as an extension of previous report<sup>11)</sup> include the physical properties of mixed asphalts, the effects of salt contents, the effects of temperature and pressure of leaching water on the leaching of radionuclides from bitumen-waste products, the effects of <sup>60</sup>Co irradiation on the pure asphalts and analysis of pure blown asphalt by infrared spectra etc.

## 2. Experimental

### (1) Materials and Equipment

a) **Apparatus:** The asphalt fixation apparatus was the same as in former studies in this laboratory.<sup>11)</sup>

b) **Composition of Wastes:** Intermediate level waste that is routinely produced at many nuclear installations is the concentrate

obtained from evaporating a composite waste made up of miscellaneous waste streams such as those from cell and equipment decontamination, solvent clean up and uranium cladding solution. This concentrate is principally a nitrate solution of sodium, potassium, aluminium and iron etc.

Accordingly, simulated waste used in this study consists of the aqueous raffinate from the second solvent extraction cycles of fuel reprocessing.<sup>12, 13)</sup>

The waste is as follows:

HNO <sub>3</sub>	3.42 M
H <sub>2</sub> SO <sub>4</sub>	0.265 M
Fe <sub>2</sub> (SO <sub>4</sub> )	0.315 M
NaNO <sub>3</sub>	1.52 M
Density	1.32 g/cm <sup>3</sup>
Specific activity	20 μCi/ml

Since acids are not compatible with asphalt, this waste was made basic with 50wt% NaOH solution before their incorporation into asphalt.

c) **Asphalt:** In this study, blown asphalt were used (supplied by Kukdong Shell Co., Ltd.) and mixed asphalts made by the ratio of straight asphalt (SA) and blown asphalt (BA) as follow:

	Ratio B A	S A	Remarks
B-0	0	100	pure straight asphalt
B-2	20	80	
B-4	40	60	
B-5	50	50	
B-6	60	40	
B-8	80	20	
B-10	100	0	pure blown asphalt

The properties of the above asphalts were measured such as softening point, flash point, penetration, ductility, density, and solubility in CCl<sub>4</sub> etc,

## (2) Procedure

### a) Preparation of bitumen-waste products:

The various asphalt were firstly melted to 150–160°C, and then 166 g of melting asphalt was charged to the evaporator in each experiment and it was continuously stirred at rates of 200 to 700 rpm while the temperature was slowly increased with the addition of waste, over a 2-hr. period, from 180–200°C. After homogenous mixing, portion of the bitumen-waste products were finally drained into specimen containers for leaching test.

b) **Leaching Test:** It has been well known for the past 15 years that leachability is another important property to characterize solidified highly radioactive waste. But, a reliable one is still missing because of the lack of a comprehensive understanding of the leaching process. Accordingly, in this study, a detailed evaluation of the leaching data on the basis of a literature review, was done by following IAEA recommendation.

Each bitumen-waste products container ( $\phi 4.5 \times 4.4$  cm) has put into leachant container ( $\phi 6 \times 12$  cm) and then 180 ml of leachant such as distilled water or sea water was poured into this container. The leachant was completely renewed with following frequency:

i) daily during the first week, ii) once a week for the following eight weeks, and iii) once a month for the following six months.

In order to evaluate the effects of temperatures and pressure of leaching water on the leaching of temperatures and pressure of leaching water on the leaching of radionuclides from bitumen-waste product, leachant container containing bitumen-waste

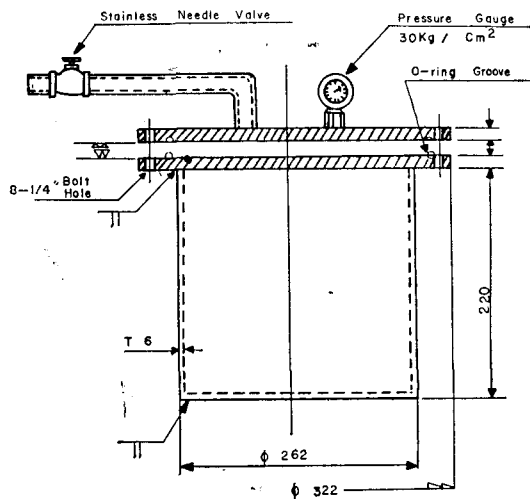


Fig. 1. Pressure Container for Leaching Test

product container was introduced into the pressure container (Fig 1) and refrigerator.

The leachate was analyzed in respect of fission products ( $^{90}\text{Sr}$  or  $^{137}\text{Cs}$ ) by G-M counter or scintillation counter.

The leaching rate was calculated from the following relation:

$$\frac{\text{activity leached/total activity in sample}}{\text{surface area of sample} \cdot \text{time}} = \frac{\text{fraction leached}}{(\text{cm}^2/\text{g}) (\text{day})}$$

c) **Radiation Test:** A major factor which must be kept in mind when one is considering the conditions for the burial of bitumen-waste products is the possible evaluation of gaseous radiolysis products. Accordingly, in order to determine the nature of the radiation-induced changes in bitumen, we carried out a number of special investigations.

Samples in the form of cylinders ( $\phi 6 \times 1.5$  cm) were irradiated in a  $^{60}\text{Co}$  facility at different dose rates, the total absorbed dose being kept virtually constant, the irradiation were performed in air.

We also determined the change of bitumen under the influence of irradiations by using infrared spectroscopy,

Table 1. Physical Properties of Mixed Asphalts

Mixed Asphalt	S-10	B-2	B-4	B-5	B-8	B-8	B-10
Specific Gravity at 25°C	1.0285	1.0314	1.0353	1.0374	1.0399	1.0450	1.0482
Penetration at 25°C 100g 5sec 1/10mm	76	52	35	28	24	17	15
Ductility at 25°C 5cm/min cm	146.5	147	90	24.5	13	5	3.5
Softening Point (B&R) °C	47.0	53.0	59.0	65.0	69.5	77.0	85.0
Flash Point °C	306	306	308	304	306	300	306
Solubility ccl <sub>4</sub> %	99.94	99.1	99.92	99.93	99.94	99.88	99.79

d) **Infrared Spectra of Asphalts:** The method employed to measure asphalt variations was based on changes in the infrared spectra of asphalt films.

In this study, procedures for the preparations of these films were followed by the method of Wright & Campbell.<sup>10</sup>

Films of asphalt were prepared by placing a small drop of molten asphalt on a sheet of cellophane film. A second sheet of unacquainted cellophane was placed over the asphalt, and the two sheets were then quickly placed in a hydraulic press in which the upper platen was maintained at approximately 120°C.

A pressure of 6000 psi was applied to the sheets and maintained for about 30 sec. The sheets were removed from the press, and the thickness of the asphalt film and two cellophane sheets was measured with a dial-thickness gage.

The films were separated from the cellophane by soaking in distilled water for several minutes, they were then mounted on aluminium panels over a 13/16 in diameter opening. After allowing the asphalt films to dry over night at room temperature, the infrared spectra of each film was recorded.

### 3. Results and Discussion

Initial experiments established that the

properties of pure mixed asphalts used for the waste incorporation process were varied by the ratio of straight asphalt and blown asphalt.

As indicated table 1, the softening point was increased with increasing the blown asphalt. It is suggested that the composition had little effect on the slight variation in flash point.

Table 1 shows that penetration was decreased with increasing blown asphalt whose penetration was lower than that of the straight asphalt.

And the properties of B-4 was especially very similar to that of Mexphalt 40/50 using typically for the incorporation of wastes into bitumen except the low ductility.

It is considered that this is due to containing 40 wt% blown asphalt whose ductility is very low.

As the direct incorporation of acidic waste into the melting asphalt hardened the asphalt, it was difficult to mix the waste with the asphalt because of its oxidation by the nitric acid in the waste. When the waste was treated with 50% solution of sodium hydroxide and became alkaline prior to bituminization, it was easy to incorporate the waste into the asphalt.

Fig. 2 represents the effects of softening point as a function of waste composition containing various salts. The softening point

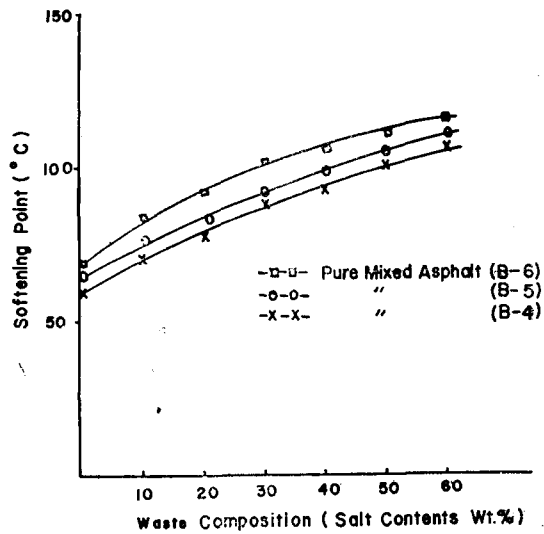


Fig. 2. Effects of Softening Point of Pure Mixed Asphalt as a Function of Waste Composition.

of bitumen-waste products(B-5) containing 40 wt% salts was 100°C and the curve in Fig 2 was shown to linear with increasing salt content.

It is considered that this is because of the increasing of viscosity of the asphalt that was attributed to the increasing of salt. Asphalt hardening was accompanied by an increased in softening point.

During the mixing for bitumen-waste products, it also seemed to be hardened the bitumen-waste mixtures with increasing salt contents.

Accordingly, it was very difficult for asphalt containing above 60% salt content to make homogeneous mixing. And also, it was not feasible to analysis to flash point of asphalts containing various salts because of production of gas and bubble by heating.

Therefore, bitumen-waste products containing 10, 20, 30, 40, 50, 60% sodium nitrate in each was implemented as an additional-tests.

As shown in Fig. 3, the flash point of asphalts was slightly lowered with incre-

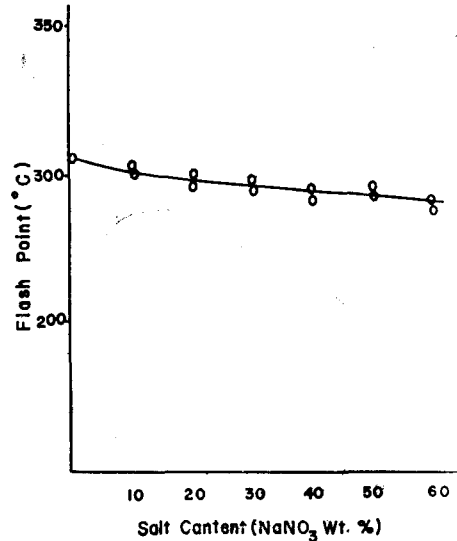


Fig. 3. Effects of Flash Point of Pure Mixed Asphalt (B-5) as a Function of Salt Content.

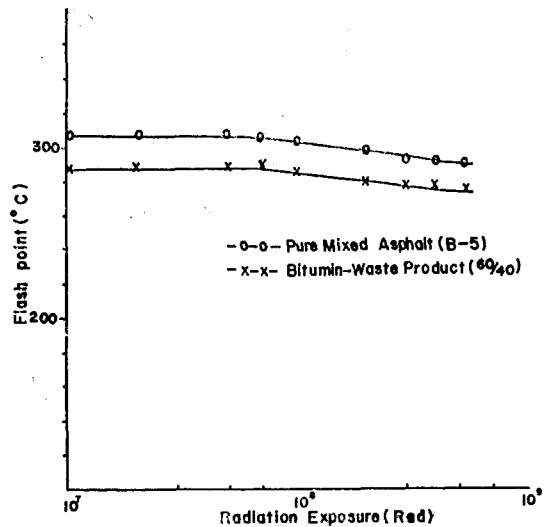


Fig. 4. Effects of Softening Point of Pure Mixed Asphalt and Bitumen-Waste Product by <sup>60</sup>Co Irradiation.

asing sodium nitrate.

The flash point of asphalts containing 60% salts was lowered by a value of 21°C, ranging from 304 to 283°C.

These results are in good agreement with studies at the Royal Military School at Br-

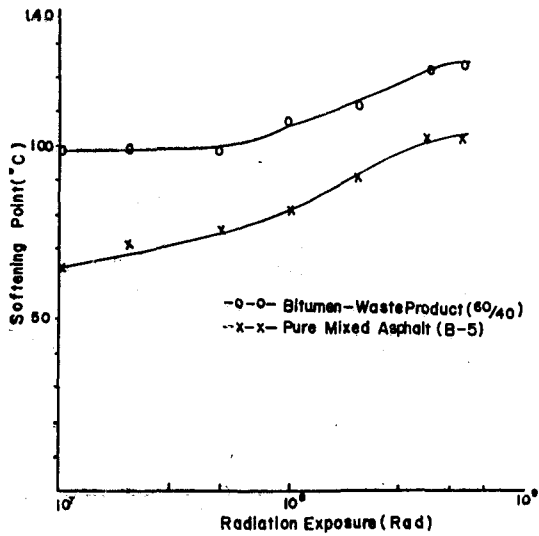


Fig. 5. Effects of Flash Point of Pure Mixed Asphalt and Bitumen-Waste Product by  $^{60}\text{Co}$  Irradiation.

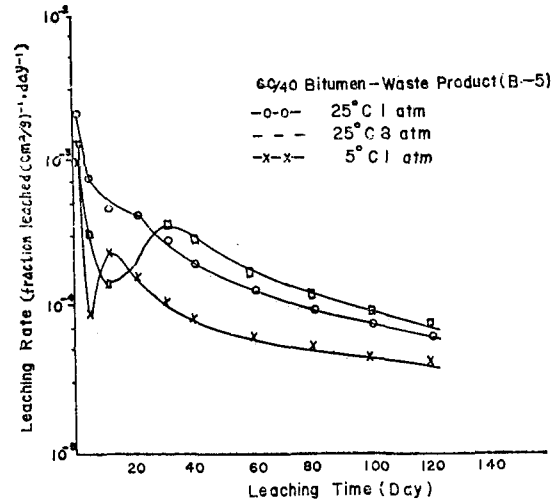


Fig. 7. Leaching Rates of  $^{137}\text{Cs}$  from Bitumen-Waste Products with Sea Water

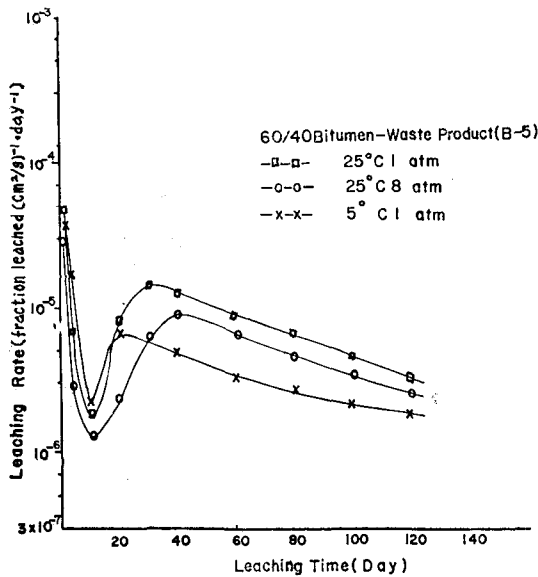


Fig. 6. Leaching Rates of  $^{90}\text{Sr}$  from Bitumen-Waste Products with Sea Water.

ussels, which reported<sup>15)</sup> that asphalt-sodium nitrate mixture were safe providing the temperature does not exceed 280°C.

The effects of softening point of pure asphalt and bitumen-waste product by  $^{60}\text{Co}$  irradiation were shown in Fig. 4. The softening point of pure mixed asphalt (B-5),

whose softening point was originally 65°C was shown to increase up to 102°C after an irradiation exposure of  $5.8 \times 10^8$  rad.

It can be probably explained that irradiation hardening was attributed to the formation of high-molecular weight compounds by irradiation. The effect of flash point of pure asphalt and bitumen-waste product by  $^{60}\text{Co}$  irradiation was shown in Fig. 5. The flash point of asphalts was slightly decreased with increasing total dose. But in both cases, the flash point is still high. Accordingly, it is considered that the slight decrease of flash point of asphalts was due to the effect to salt contents rather than that of  $^{60}\text{Co}$  irradiations. The volume changes of bitumen-waste products as a function of radiation exposure with increasing salt content was follows:

By Rodier,<sup>3, 4)</sup> it was reported that sample of Mexphalt 40/50 and R 90/40 irradiated to a dose of  $10^9$  rad showed increases of 74% and 25% in volume, respectively, but each

product with Mevphalt 40/50 and with R 90/40 containing 57.2 wt% salts showed a increased of 28.6% and 0% in volume, respectively.

All of the samples made with mixed asphalt used in this study were irradiated at the intensity of 32 rad/sec and 12 rad/sec. At a total dose of  $5.8 \times 10^8$  rad up to the present, irradiation of these samples shows no evidence of volume and cause no swelling.

It is generally known that during irradiation, asphalt evolves gas, principally hydrogen and hydrocarbons and small amounts of carbon dioxides, and the evolution of gas produces on increase in volume of the asphalt<sup>16)</sup>, but the asphalt investigated in this study is unaffected by an irradiation of  $5.9 \times 10^8$  rad.

Accordingly, it is considered that this is attributed to the effect that the forms are more porous and radiolytic gases can escape.

Therefore, compared with other results, the asphalts used in this study are considered to be satisfactory from radiation stability.

When bitumen-waste products dumped into deep sea for ultimate disposal, they would remain on the sea bottom for an extremely long period of time at the high pressure and low temperature of the deep sea water. Therefore, it is certainly of significant to learn what influence these conditions would have on the leaching characteristics of the bitumen-waste products.

Bitumen-waste products were subjected to leaching tests with artificially prepared sea water at 8 atm.

Fig. 6 and 7 shows the relationship between the leaching rate and leaching time.

As indicated Fig. 6 and 7 the fraction of  $^{90}\text{Sr}$  and  $^{136}\text{Cs}$  leached for periods of 120 days was  $2.1 \times 10^{-6} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$ ,  $6.02 \times 10^{-5} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$ , respectively. The fraction of  $^{90}\text{Sr}$  was much lower than that of  $^{137}\text{Cs}$ .

The leaching rates of both nuclides decrease as the pressure increase.

This suggests that there is a correlation between the decrease in volume indicated by changes of strain due to the compression and the decrease in leaching rate, that is, a possibility that the decrease in volume shows up the diffusion process within the water held in voids in the bitumen-waste product. As this would reduce the contamination of sea water, this decrease in leaching rate under high pressure may be regarded as favouring deep sea dumping.

The leaching rate shown in Fig. 6 and 7 show a clear tendency to a sudden increase in the test range from one week to two weeks. It is interesting to note that this tendency is attributed to the salt content in the surface of bitumen-waste product.

Fig. 6 and 7 give the results of leaching tests on a bitumen-waste products containing  $^{90}\text{Sr}$  or  $^{137}\text{Cs}$  tested at  $5^\circ\text{C}$  and atmospheric pressure, together with bitumen-waste products of the same composition tested at  $25^\circ\text{C}$  and the same pressure. The fraction of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  leached for periods of 120 days at  $5^\circ\text{C}$  was  $1.7 \times 10^{-5} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$  and  $4.01 \times 10^{-5} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$ , respectively.

As shown in Fig. 6 and 7, the leaching rate exhibited a difference of both products at a stage of leaching for periods of 120 days. The leaching rates of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  were much lower than those in room temperature.

But the leaching rate of both samples



**Table 2. Effects of Pressure and Temperature of Leaching Water on the Leaching of <sup>90</sup>Sr from Bitumen-Waste Products with Distilled Water**

Condition	Material to be mixed		Leaching Rate [fraction leached (cm <sup>2</sup> /g) <sup>-1</sup> day <sup>-1</sup> ]						
	SA	BA	1d	7d	20d	40d	80d	100d	120d
Pressure (8 atm)	60	40	3.51×10 <sup>-5</sup>	6.98×10 <sup>-7</sup>	1.39×10 <sup>-5</sup>	1.12×10 <sup>-5</sup>	3.21×10 <sup>-6</sup>	3.15×10 <sup>-6</sup>	2.92×10 <sup>-6</sup>
	50	50	6.31×10 <sup>-5</sup>	1.29×10 <sup>-6</sup>	1.69×10 <sup>-6</sup>	1.39×10 <sup>-5</sup>	5.13×10 <sup>-6</sup>	4.65×10 <sup>-6</sup>	4.65×10 <sup>-6</sup>
	40	60	1.32×10 <sup>-4</sup>	3.32×10 <sup>-6</sup>	8.45×10 <sup>-6</sup>	2.25×10 <sup>-5</sup>	9.12×10 <sup>-6</sup>	7.32×10 <sup>-6</sup>	5.63×10 <sup>-6</sup>
Temperature (5°C)	60	40	3.22×10 <sup>-5</sup>	8.51×10 <sup>-7</sup>	7.12×10 <sup>-7</sup>	5.91×10 <sup>-6</sup>	1.52×10 <sup>-6</sup>	1.14×10 <sup>-6</sup>	1.05×10 <sup>-7</sup>
	50	50	4.9×10 <sup>-6</sup>	1.52×10 <sup>-6</sup>	1.18×10 <sup>-5</sup>	7.12×10 <sup>-6</sup>	2.22×10 <sup>-6</sup>	2.22×10 <sup>-6</sup>	1.95×10 <sup>-6</sup>
	40	60	1.49×10 <sup>-7</sup>	3.75×10 <sup>-6</sup>	1.75×10 <sup>-6</sup>	1.32×10 <sup>-5</sup>	1.22×10 <sup>-6</sup>	3.13×10 <sup>-6</sup>	2.57×10 <sup>-6</sup>

SA: Straight Asphalt BA: Blown Asphalt

**Table 3. Effects of Pressure and Temperature of Leaching Water on the Leaching of <sup>137</sup>Cs from Bitumen-Waste Products with Distilled Water**

Condition	Material to be mixed		Leaching Rate [fraction leached (cm <sup>2</sup> /g) <sup>-1</sup> day <sup>-1</sup> ]						
	SA	BA	1d	7d	20d	40d	80d	100d	120d
Pressure (8 atm)	60	40	6.52×10 <sup>-4</sup>	8.52×10 <sup>-5</sup>	1.41×10 <sup>-4</sup>	9.02×10 <sup>-5</sup>	8.52×10 <sup>-5</sup>	7.16×10 <sup>-5</sup>	5.67×10 <sup>-5</sup>
	50	50	1.49×10 <sup>-3</sup>	2.43×10 <sup>-4</sup>	2.20×10 <sup>-4</sup>	2.35×10 <sup>-4</sup>	1.34×10 <sup>-4</sup>	9.21×10 <sup>-5</sup>	7.92×10 <sup>-5</sup>
	40	60	1.98×10 <sup>-3</sup>	3.51×10 <sup>-4</sup>	7.42×10 <sup>-4</sup>	3.62×10 <sup>-4</sup>	2.16×10 <sup>-4</sup>	1.28×10 <sup>-4</sup>	8.55×10 <sup>-5</sup>
Temperature (5°C)	60	40	4.51×10 <sup>-4</sup>	1.05×10 <sup>-4</sup>	6.10×10 <sup>-5</sup>	2.05×10 <sup>-5</sup>	1.72×10 <sup>-5</sup>	1.23×10 <sup>-5</sup>	1.13×10 <sup>-5</sup>
	50	50	1.42×10 <sup>-3</sup>	1.42×10 <sup>-4</sup>	2.82×10 <sup>-4</sup>	8.09×10 <sup>-5</sup>	4.15×10 <sup>-5</sup>	4.02×10 <sup>-5</sup>	3.92×10 <sup>-5</sup>
	40	60	5.95×10 <sup>-3</sup>	7.52×10 <sup>-4</sup>	5.11×10 <sup>-4</sup>	1.41×10 <sup>-4</sup>	4.97×10 <sup>-5</sup>	4.86×10 <sup>-5</sup>	4.42×10 <sup>-5</sup>

SA: Straight Asphalt BA: Blown Asphalt

turn out to be approximately the same after leaching for periods of 120 days.

Accordingly, it appears that these results could be influenced leaching rate by low temperature and high pressure, and saved by improving safety in the dumping of sea.

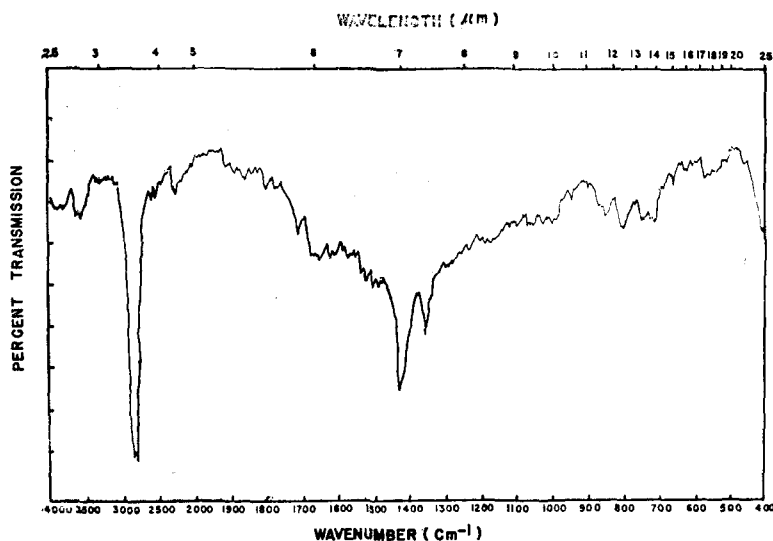
For the effect of leachants from the standpoint of leaching rates, bitumen-waste products also were implemented the leaching test with distilled water at the high pressure and low temperature. The results were presented in Table 2 and 3. However, no difference in the leaching rate with sea and distilled water was observed for the bitumen-waste

product containing 40 wt% salts.

And also, the effect of leaching rate on the mixed asphalt was shown that leaching rate was slightly decreased with increasing straight asphalt. It is considered that this is due to straight asphalt to be less permeable than blown asphalt when compared at equivalent hardness.<sup>17)</sup>

Accordingly, diffusion of the water molecules within the bitumen-waste products seems to be the factor controlling permeation rate rather than the rate at which water is absorbed.

The infrared spectra of asphalt films showed a marked similarity in their absorption characteristics.



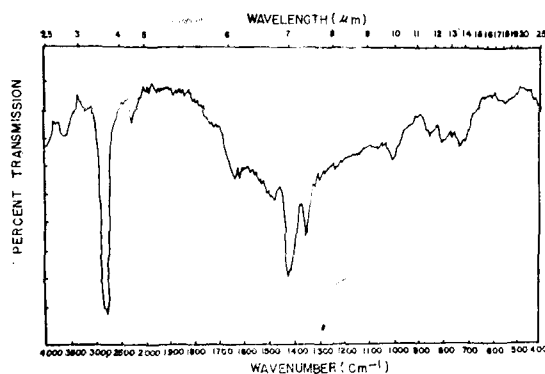
**Fig. 8. Infrared Spectra of Blown Asphalt (25 $\mu$  thick)**

The asphalts had absorption bands at the same or nearly the same wavelengths and their spectra differed primarily in the transmittance values of some of these bands. The structural grouping which appeared to be present in the asphalts examined were hydroxyl,  $-\text{CH}_2-$ ,  $\text{C}-\text{CH}_3$ , carbonyl,  $-(\text{CH}_2)_4$  and aromatic rings. There were also absorption bands which indicated aromatic substituents or fused rings. Accordingly, the infrared spectra of blown asphalt used in this study was shown in Fig. 8 and 10.

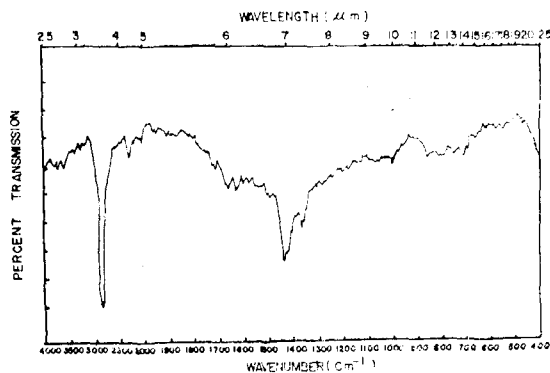
The structural assignments represent in the form of O-H group absorbing at 2.78  $\mu$ ,  $-\text{CH}_2-$  and  $\text{C}-\text{CH}_3$  groups absorbing at 3.58  $\mu$  and 6.97  $\mu$ ,  $\text{C}-\text{CH}_3$  group absorbing at 7.27  $\mu$ .

There are in agreement with the fact that most of the asphalt fractions absorb near 3-7.2  $\mu$  because of the stretching and angle-deformation vibrations of the  $\text{CH}_2$  and  $\text{CH}_3$  groups of alkanes.<sup>10)</sup>

But it is difficult to interpret for a band found at 4.5  $\mu$ . It is considered that this is probably due in part to  $-\text{CH}_2-$  chains because there are certainly aliphatic  $\text{CH}_2$



**Fig. 9. Infrared Spectra of Blown Asphalt After  $^{60}\text{Co}$  Irradiation (25 $\mu$  thick)**



**Fig. 10. Infrared Spectra of Blown Asphalt (100 $\mu$  thick)**

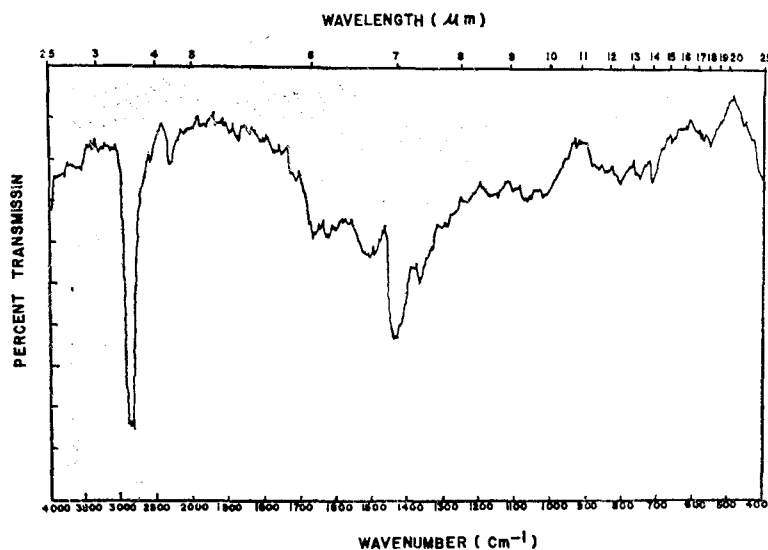


Fig. 11. Infrared Spectra of Blown Asphalt after  $^{60}\text{Co}$  Irradiation ( $100\ \mu$  thick)

and  $\text{CH}_3$  group present.

According to other workers,<sup>14, 18)</sup> significant changes in absorbance are particularly measured at  $5.88\ \mu$  ( $1700\ \text{cm}^{-1}$ ),  $6.25\ \mu$  ( $1600\ \text{cm}^{-1}$ ). Respective absorbances at these wavelengths are due to  $\text{C}=\text{O}$ , aromatic and conjugated alkene banding.

It was known that the carbonyl band was relatively free from interference by absorbances of other groups and carbonyl absorbances can be determined with greater accuracy than other groups such as  $\text{OH}$  and  $\text{C}-\text{O}$  and this band was used extensively by the authors in previous investigations on the infrared spectra of asphalt, but the identification of the types of organic molecules responsible for the absorbance at  $5.88\ \mu$  ( $1700\ \text{cm}^{-1}$ ) was not made.

However, according to the data studied in this experiment, the absorbance changes at  $5.88\ \mu$  ( $1700\ \text{cm}^{-1}$ ) does not coincide with the band found in this region in the spectrum of other workers. Aromatic substitutions are also observed at  $11.8\ \mu$ ,  $12.5\ \mu$  and  $13.7\ \mu$ .

The effect of film thickness of infrared

spectra is observed in Fig. 8 and 10.

Infrared spectra of thick ( $100\ \mu$ ) asphalt films (Fig. 10) was nearly the same as that which took place in the thin ( $25\ \mu$ ) films (Fig. 8).

The infrared spectra of films of asphalts irradiated at a total dose of  $5.8 \times 10^8$  rad up to the present was shown in Fig. 9 and 11. These results indicate that the functional groups of blown asphalt are not nearly affected by irradiation.

However, the strong band found at  $10\ \mu$  ( $1000\ \text{cm}^{-1}$ ) in Fig. 9 was puzzling, it is considered that it occurs in a region where certain  $\text{C}-\text{O}$ ,  $\text{S}=\text{O}$ ,  $\text{Si}-\text{O}$  and naphthenic groups absorb.<sup>19)</sup>

#### 4. Conclusion

The principal conclusion investigated in this study is as follows.

1) Incorporation of wastes in bitumen has proved to be a very desirable method of fixing radioactive wastes in a relatively insoluble matrix. The physical properties of

bitumen-waste products have shown that the softening point of bitumen-waste product(B-5) was linear with increasing salt contents and the flash point of asphalts was slightly lowered with increasing sodium nitrate.

2) The effect of physical properties of pure asphalt and bitumen-waste products by  $^{60}\text{Co}$  irradiation was shown that the softening point of pure asphalt increases up to  $102^\circ\text{C}$  at a total dose of  $5.8 \times 10^8$  rad.

At a total dose of  $5.8 \times 10^8$  rad, up to the present, irradiation of asphalts showed no evidence of volume and caused no swelling.

3) The effects of temperature and pressure of leaching water on the leaching of radionuclides from bitumen-waste products containing 40% salts have shown that the fraction of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  leached for periods of up to 120 days at 8 atm was  $2.1 \times 10^{-6} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$  and  $6.02 \times 10^{-5} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$  respectively, and at  $5^\circ\text{C}$   $1.7 \times 10^{-7} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$  and  $4.01 \times 10^{-5} (\text{cm}^2/\text{g})^{-1} \text{ day}^{-1}$ , respectively. These values were slightly lower than those in atmospheric pressure. But the effect of temperature was much lower than those in room temperature.

4) No difference in the leaching rate with sea and distilled water was observed for the bitumen-waste products containing 40 wt% salts. Chemical and physical properties derived from the infrared spectra of asphalt fractions can be summarized that blown asphalts are predominantly branched-chain aliphatics with slight aromatic content, and the functional groups of blown asphalt were not nearly affected at a total dose of  $5.8 \times 10^8$  rad up to the present, and also variations in film thickness are not observed.

Further studies on the infrared spectra of asphalts is being demonstrated in detail.

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