Combustible Properties of LLW Bitumen Packages in a Fire Test

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BACKGROUND

Most Low Level Wastes derived from Japanese nuclear power stations are classified as LSA-II・III material according to ss6 §131 of the IAEA '85 regulations, while whole packages composed of wastes and packaging are classified as Industrial package according to ss6 §131 of the regulations. Concerning LSA material, the fire test for Type B package is not regulated, because a contained activity is limitted by the A2 value itself. According to the regulation of the Fire Defense Agency in Japan, straight asphalt is classified as "semi-dangerous material" because of its calorific power; bitumen package is classified as normal material because of its lower calorific power. However, among these LLW wastes, test resuls have indicated that bitumen packages are flammable.

In light of this situation, a fire test for bitumen packages with some simulated nuclides was performed by CRIEPI in 1988 and 1989 in order to discover whether bitumen packages are combustible and also to determine how many nuclides contained in the packages are released into the environment in the event of a fire accident.

METHODS

Specimens

Two types of specimens were provided in the test. Small beaker-scale samples were made for the purpose of studying the ignition temperature, the state of burning, and unification by mixing and finally researching the way aerosol is sampled.

The actual size specimens contained in 200 liter drums (by JIS standard Z1600) were created for the purpose of examining the state of burning generally, specifically, the ignition temperature, the temperature distribution, combustion characteristics, the emissivity of surface, the quantity of released nuclides, and the nature of analyzing the residue. The volume of both specimens was about 100cc and 160~170 liter.

The primary materials used to create the specimens were 60/40 grade **straight asphalt** (bitumen) by the JIS standards and some chemicals (**Co**, **Sr**, **Cs**, and **Eu**, and **$NaBO_2 \cdot 1/2H_2O$**). When selecting nuclides, we referred the nuclide composition of bitumen packages derived from PWR power plants in Japan. In addition, Eu was used as a substitute of α nuclides because of the similarity of its chemical characteristics. The mixing ratio of chemicals and asphalt is described in **Table 2**. The ratio was decided in the preliminary test after consideration of the sensitivity of detectors. As a result, the ratio is rather high in comparison to real LLW packages.

Table 1. The description of Specimens

Specimens	Volume	Test items
Beaker-scale	about 100cc	Ignition temperature Uniformity of wastes Combustion characteristics Way of aerosol sampling
Actual size 200 Lit. drum JIS Z1600	about 160 lit. ~ 170 lit.	Ignition temperature Temperature distribution Combustion characteristics Emissivity of surface Analysis of generating gas Analysis of the residue

Table 2. The ratio of additives

Additive	Ratio	Note
Co,Sr,Cs	0.2wt%	of total weight (as sulfate)
Eu	0.2wt%	of total weight (as oxide)
$NaBO_2 \cdot 1/2H_2O$	Waste/Asphalt = 40/60	40wt% of total weight

Test Methods

The small samples were heated in an electric furnace along the ISO heating curve, and the actual size specimens were heated in the large furnace in the same manner. The ISO heating curve was set to raise the ambient temperature from 30℃ to 835℃ for 35min. as described in IAEA '72 Regulation §727. An example of ISO heating curve is shown in **Fig.1**.

When heating specimens, we used the devices shown in **Fig.2** for beaker-scale specimens, and the large directly heating furnace shown in **Fig.3** for actual size specimens. The main specifications of the furnace are as follows :

Type of furnace : Direct heating on moving truck
Inside measurement : 5W x 5H x 8.5L (m)
Heating range : 600 ~ 1200℃
Fuel : LPG

The manner in which the resolved gas was collected is indicated in **Fig.2** and **Fig.4**. As shown in those figures, the resolved gas was collected by a filter holder or gas collector, cooled, and then sampled.

As shown in **Table 1**, the major tasks in the test were measurement of the state of burning, temperature distribution, and the temperature of ignition and collection and analysis of aerosol and analysis of the residue. In addition, emissivity of the surface of some packages was measured.

For the actual size specimen, it was confirmed that the total heat input from the ISO heating curve was almost equal to the heat input from the fire test for the Type B package, when compared by the thermal analysis code 'CRISCAT' which was developed by CRIEPI. The specifics of the CRISCAT code are detailed in another CRIEPI report offered at this conference.

RESULTS

Temperature and Burning

1) When the ambient temperature reached about 600℃, the top surface temperature (No.1) of the asphalt package without hood rose to the 350 ~ 380℃ range and resolved gas derived from the asphalt package ignited automatically. At the high temperature region (about 800℃) of the ISO heating curve, the side surface temperature (No.2) remained in the 320 ~ 430℃ range. At the same region of the curve, the temperature of a point 20mm away from the top surface (No.3) came close to the surface temperature just after the burners were shut off.

2) While heating specimens with a hood, the maximum temperature of the center of actual size specimens was about 50℃, and also the maximum temperature of points 60mm away from each surface was about 70℃, that is, most of the specimen, including the center, did not melt because of its large heat capacity. In contrast, for specimens without a hood (an opened drum), the center region's temperature rose to the 150 ~ 200℃ range. By the end of the heating procedure the specimen had melted completely. Here, the melting temperature of asphalt is about 120℃.

3) While burning, the bottom of the flame was located 10cm away from the upper surface of specimens. This phenomenon indicates that the surface of the specimens did not burn, but that the resolved gas which had been heated in the ascending current did. When the heating had been completed, specimens with a hood extinguished the fire automatically; specimens without a hood (an opened drum) kept on burning for 12 hours after the end of the heating procedure. In order for such extended burning to take place, both excessive heat and sufficient oxygen must have been provided continuously.

4) Measured emissivity of specimens rose from 0.77 to 0.96 while the surface temperature of specimens rose from 640℃ to 680℃. It is supposed that the deterioration of painting and oxidation of painting or steel affected the change of emissivity.

Analysis of Gas

1) Results of the beaker-scale test revealed that the resolved gas was composed of a wide range hydrocarbon (H.C.), including $C_1 \sim C_{40+}$ and H_2O, and at

450℃ the gas included C_{40+}H.C. at a rate of 40%. The volume of the resolved gas was about 27 KLit./kg.

2) According to the analysis of resolved gas, the volatile ratio of Cs was greater than other three nuclides because of its typical volatility. The details of volatility data were as follows.

Table 3. The volatile ratio of nuclides

Nuclides	Volatile ratio
Co, Sr, Eu	$1.9 \times 10^{-3}\% \sim 3.6 \times 10^{-4}\%$
Cs	$1.5 \times 10^{-2}\% \sim 4.7 \times 10^{-3}\%$

* Volatile ratio
= (released nuclide)/(total additive of each nuclide)

The residue

1) According to the results of the chemical analysis of boring samples, which had been pulled out through 6 plugs on the side of specimens, the ratio of waste (added chemicals:$\mathbf{NaBO_2 \cdot 1/2H_2O}$) was greater in the upper region than in other areas.

2) Similarly, Cs and Eu tended to concentrate in the upper region and the softened region near the side wall. Co tended to concentrate in the colder region including the center and in the upper region. Further investigation will be necessary in order to fully explain this phenomenon, but we believe that the movement of the generated gas into the softened region and the convection of fluid asphalt concerned this phenomenon.

3) The loss of weight was about 10% in the case of the semi-shielded drum (with a hood), and about 20% in case of the non-shielded drum (an opened drum).

CONCLUSION

1) When the surface temperature of specimens rises to the 350 ~ 380℃ range in a fire environment, the resolved gas will be generated and ignite automatically. In order to keep on burning, it is necessary to supply both excessive heat and sufficient oxygen.

2) The volatile ratio of Cs, which is a typical volatile nuclide, is greater than other nuclides. However, its value is 1.5 wt% of added sulfate at the maximum.

REFERENCES

ISO Recommendation R834 "Fire Resistance Tests of Structures", 1963

Wataru,M. "Development of Heat-Transfer and Thermal Analysis Codes for Radioactive-Material Packages (CRISCAT)" PATRAM'92 (1992)

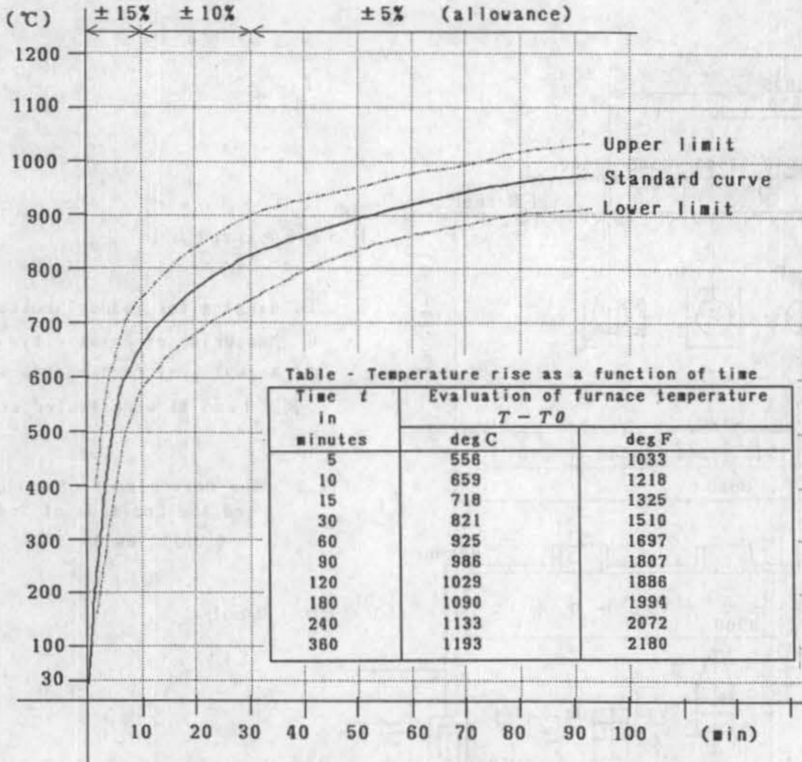


Table - Temperature rise as a function of time

Time *t* in minutes	Evaluation of furnace temperature $T - T0$	
	deg C	deg F
5	556	1033
10	659	1218
15	718	1325
30	821	1510
60	925	1697
90	986	1807
120	1029	1886
180	1090	1994
240	1133	2072
360	1193	2180

Fig.1 I S O standard temperature rise curve

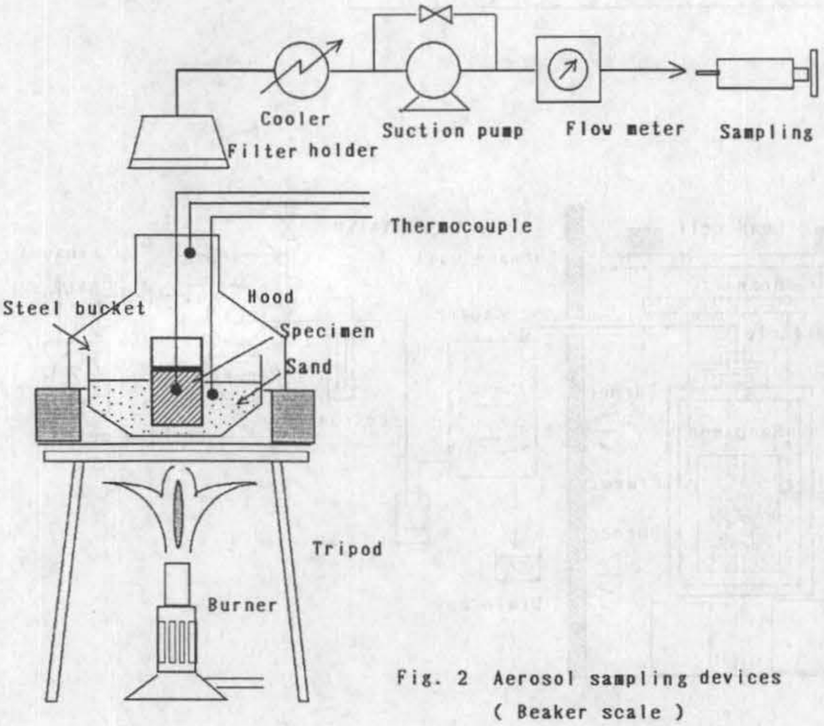
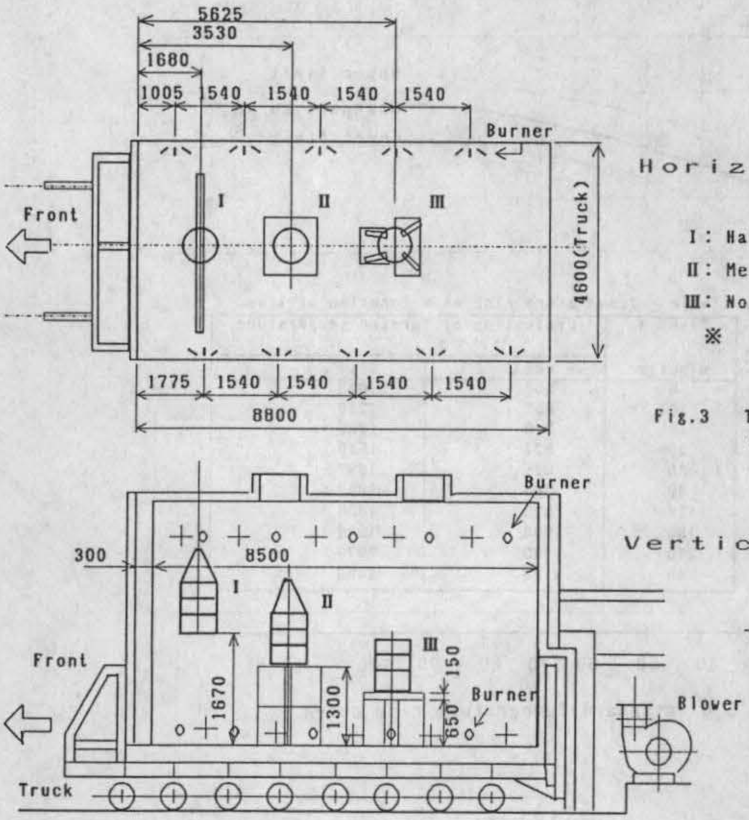


Fig. 2 Aerosol sampling devices
(Beaker scale)



Ⅰ: Hanging for gas collection
Ⅱ: Measuring of Emissivity
Ⅲ: Normal test (set on blocks)
※ Ⅰ and Ⅲ were tested at the same tim

Fig.3 The Measurement of Furnace and the Position of Specimens (unit: mm)

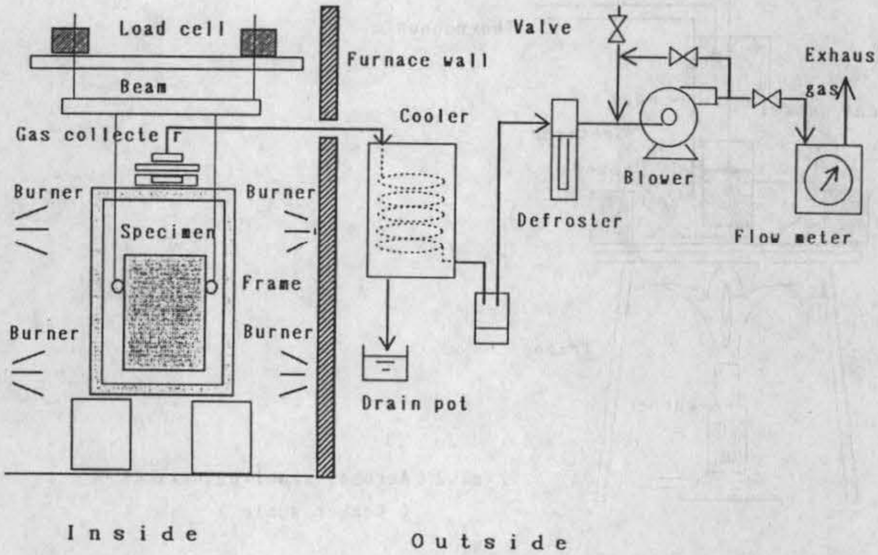


Fig. 4 Aerosol sampling devices (Actual size)

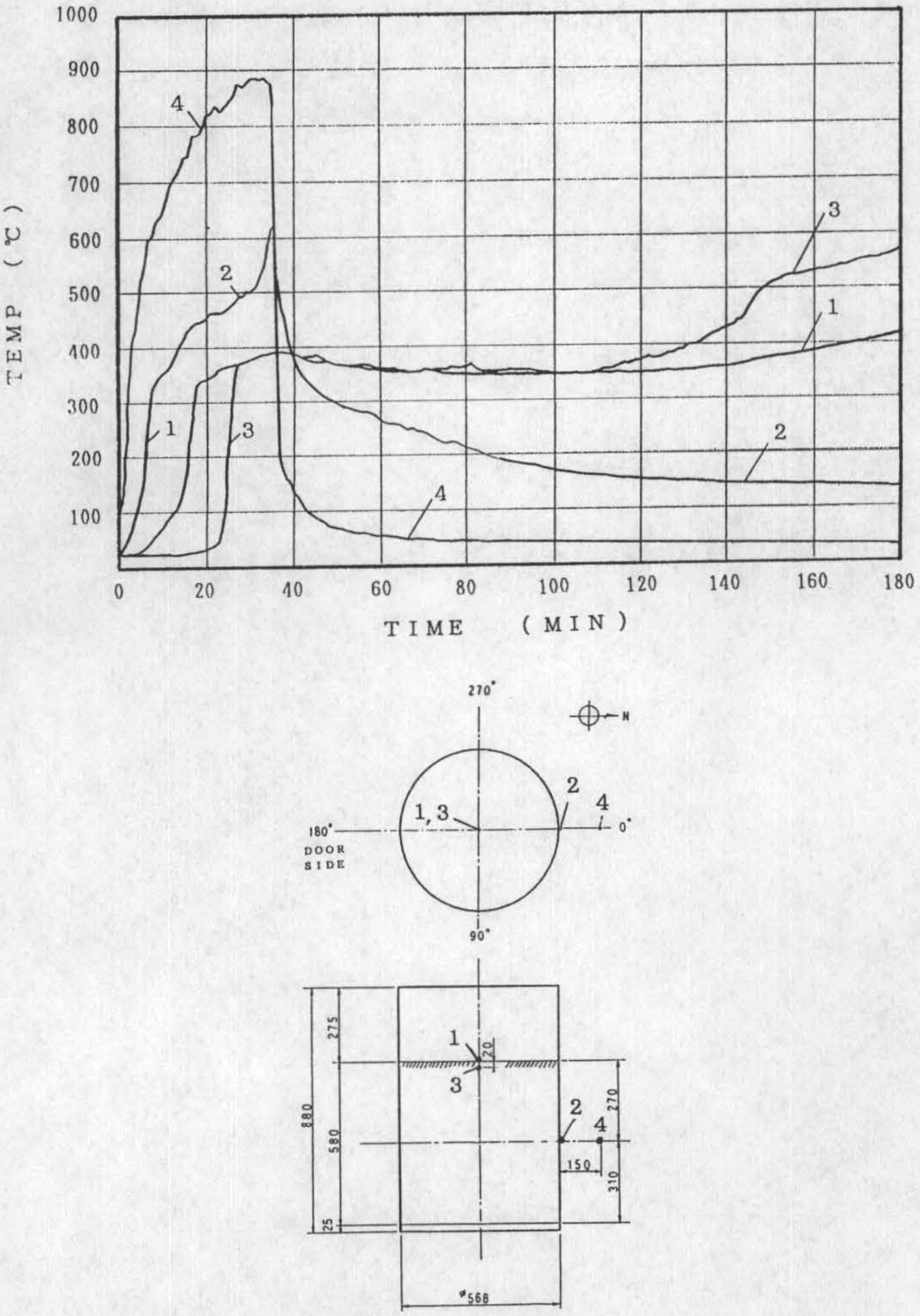


Fig.5 TEMPERATURE HISTORY UNDER TESTING

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