

Diffusion and Vapour-Rise of Methane Vapour from Areal Source in Air Stream

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(Received September 10, 1968)

Introduction

Methane has become an important fuel in industry, and its transportation and storage on a large scale in its liquid phase have been realized. If the liquid methane of large quantities is flowed out by an unexpected accident, it is a very serious problem, how the methane vapour will be diffused.

Concerning vapour-rise and diffusion, there are some reports,¹⁾ but chemical engineers are apt to treat them only in closed spaces. Of course, these phenomena in closed spaces, such as buildings of factories, have important significances. However, the results obtained in closed spaces are not adequate in the open air. In the open air, the wind always blows, and in even very light wind, such as 0.5 m/sec, phenomena are very different compared with those in closed spaces. For example, diffusion coefficient (turbulent diffusivity) in the open air should be considered as $10^5 \sim 10^6$ times as large as that (molecular diffusivity) in closed vessel.

As there have been scarcely any reports concerning vapour-rise and diffusion in air stream, we carried out some experiments in a wind tunnel, even though they were small scale experiments.

Effects of density of methane gas

Relative densities of methane gas referred to air are shown in Table 1. Methane gas has the tendency to ascend, owing to its small relative density at room temperature, but generally this tendency is often overestimated.

If gas is mixed with air, the resulted relative density ρ_{mix} can be calculated by next formula:

$$\rho_{\text{mix}} \doteq 1 - c(1 - k) \quad (1),$$

where k is the relative density referred to air and c is the volume concentration of gas in air. The relative density k of methane at room

Table 1. Relative density of methane vapour referred to air.

Temperature (°C)	Relative Density
-160	1.47
-113	1.00
0	0.59
15	0.56

Table 2. Mixed relative density and vapour concentration.

Concentration (%)	Mixed relative Density
100	0.56
30	0.89
23	0.90
10	0.96

temperature is 0.56, so the values of the resulted relative densities for some concentrations are shown in Table 2, and we can recognize that even very light gas as methane, the mixed density becomes only 0.90, even in considerably dense state such as 23%.

Furthermore, immediately after the liquid vaporized, the vapour temperature is still low, and k is rather near to 1 (Table 1), so the mixed density should be considered much larger than that considered above.

Procedure of experiment

We used methane vapour, and also used di-ethyl-ether vapour as contrast. A flat plate was set in the wind tunnel in Ochanomizu University, whose test section is 50 cm \times 50 cm in cross section and 200 cm in length, and in a certain part of the plate, a block of poly-styrene foam was attached. In a portion of the block, a dish-shaped cavity was formed, and a dish made by poly-vinyl-chloride was put in the cavity, whose depth was 3.5 cm and the inner volume of the cavity was 88 cc. In order to make the flow turbulent, an L-shaped metal, the length of its edge was 19 mm, was set at the leading edge of the plate. A gas sampling probe was set at one position, 30.75 cm leeward (x -ward) from the center of the cavity, but it was movable in vertical direction (z -direction). The concentration of the sampled gas was measured by a hydrogen-flame-ion detector of a gas chromatograph and recorded by a recorder. The wind speed in these experiments was 1 m/sec. (Fig. 1)

Liquid methane or ether was poured directly into the cavity (Case A), or they were poured into the cavity in which water had been filled up to the depth of 1.75 cm (Case B₁ for methane and B₂ for ether).* Except the case B₁, the probe was moved upward by a travelling device at the speed of 1 mm per 3.3 sec., so the gas concentration was measured continuously. In the case B₂, as the evaporating rate of methane was very rapid, we fixed the probe at several heights,

* The air temperature was about 32°C and the water temperature was about 30°C.

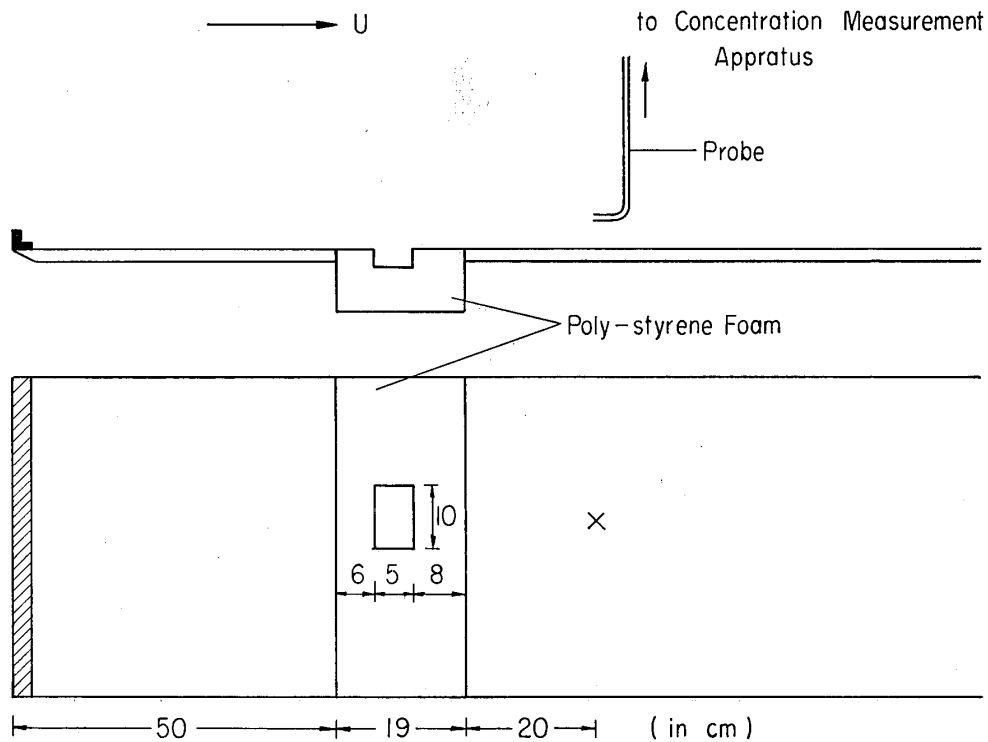


Fig. 1. Arrangement of equipments.

and recorded the concentration during several seconds, and for each measurement, water and liquid methane were renewed.

Results

The results in the case A are shown in Fig. 2 (i) and (ii), and those in the case B are shown in Fig. 3 (i) and (ii). The concentration profiles show clearly that the peak concentration appears on the plate surface and the profiles take the form

$$C \propto \exp(-z/B) \quad (2)$$

for both gases, methane and ether, in spite of considerable difference between their densities.

If gas ascends owing to its low density, the pattern of gas plume would be that shown in Fig. 4 (A), just like the smoke plume emitted from a stack in the light wind, and the position of the peak concentration would be at some height above the surface (Fig. 4 (B)); the experimental results, however, show that the peak concentrations appear closely near the surface.

According to the work of one of the authors,²⁾ the concentration can be calculated by next formula:

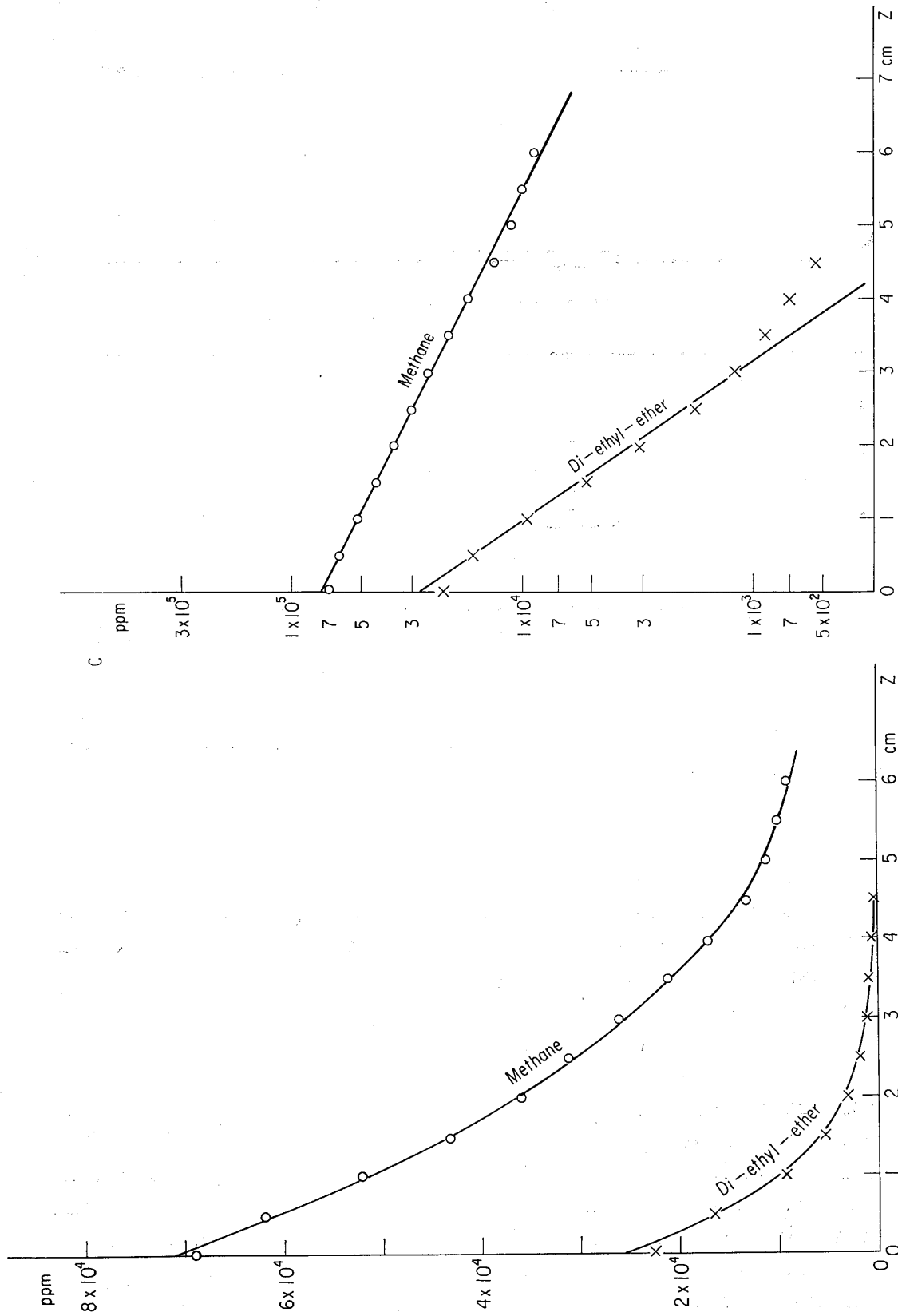


Fig. 2 (i).
Vertical concentration profiles of methane or di-ethyl-ether (poured directly into the vessel).
Fig. 2 (ii).

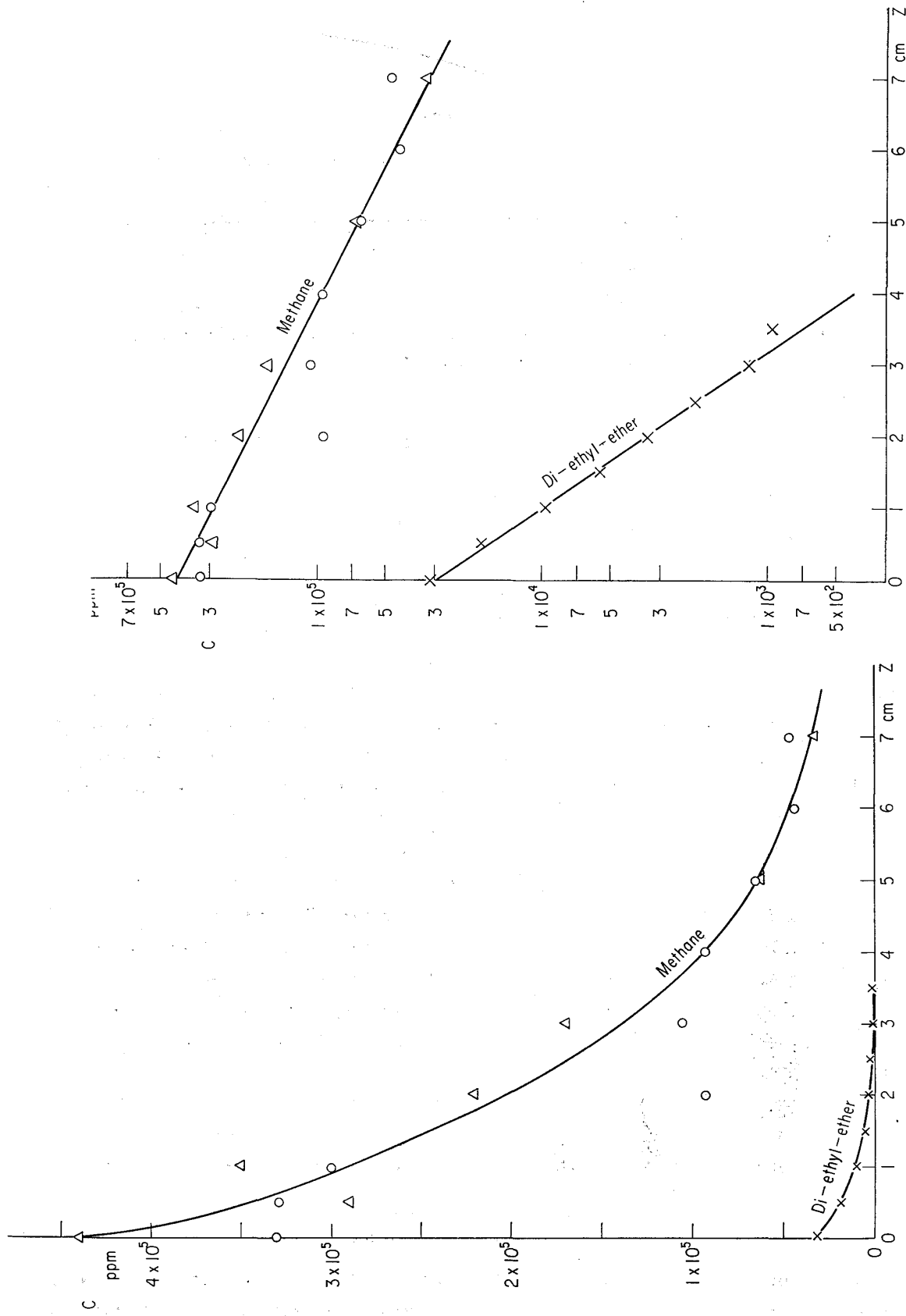


Fig. 3 (i).
Vertical concentration profiles of methane or di-ethyl-ether (poured upon water surface).

Fig. 3 (ii).

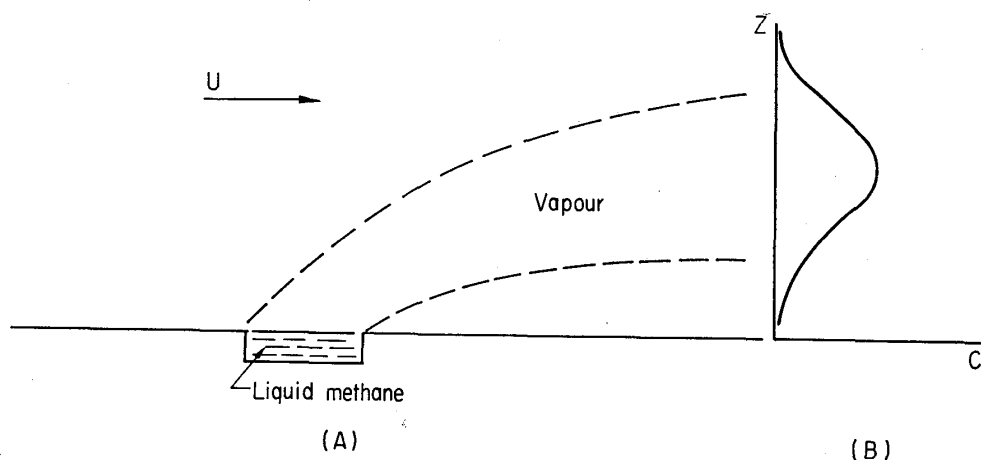


Fig. 4 (A). Fictitious pattern of vapour evaporated from liquid surface.
 (B). Fictitious vertical concentration profile of vapour.

$$C \doteq \frac{q}{u} \frac{e^{-\frac{z}{B}}}{2} \frac{\sqrt{A}}{2} \left[\Lambda \left(\frac{x+l}{\sqrt{A}} \right) - \Lambda \left(\frac{x-l}{\sqrt{A}} \right) \right] \quad (3),$$

$$A = q_A(\varphi_A x + e^{-\varphi_A x} - 1) \quad (4),$$

$$B = q_B(\varphi_B x + e^{-\varphi_B x} - 1) \quad (5),$$

$$\Lambda(\xi) = \xi \operatorname{erf}(\xi) + \frac{1}{\sqrt{\pi}} (e^{-\xi^2} - 1) \quad (6),$$

where q is the rate of evaporation per unit area, x is leeward distance from the center of the cavity, and l is a half length of the cavity. The formula shows that the vertical concentration profile is proportional to $\exp(-z/B)$, which coincides with the experimental results.

The measured values of B are 2.70 for methane vapour and 0.94 for ether vapour respectively, and the latter co-incides with the results obtained by the authors for line source³⁾ and point source.⁴⁾ The value of B for methane vapour is about 3 times as large as that for ether. This difference is due to the fact that the liquid methane at very low temperature (-164°C) is poured into the vessel or upon the water surface at room temperature (30°C ca.), and the liquid methane evaporates vigorously and the liquid surface was agitated violently, so the turbulent diffusivity became much larger and the value of B became larger.

In these experiments, the available liquid methane was not sufficient, so we could not measure at several leeward positions; therefore, we could not calculate the concentration by the formule (3)~(6).

Rate of vaporization

We wished to know the order of magnitude of the rate of vaporization, we measured the decrease of weight of methane in a thermally

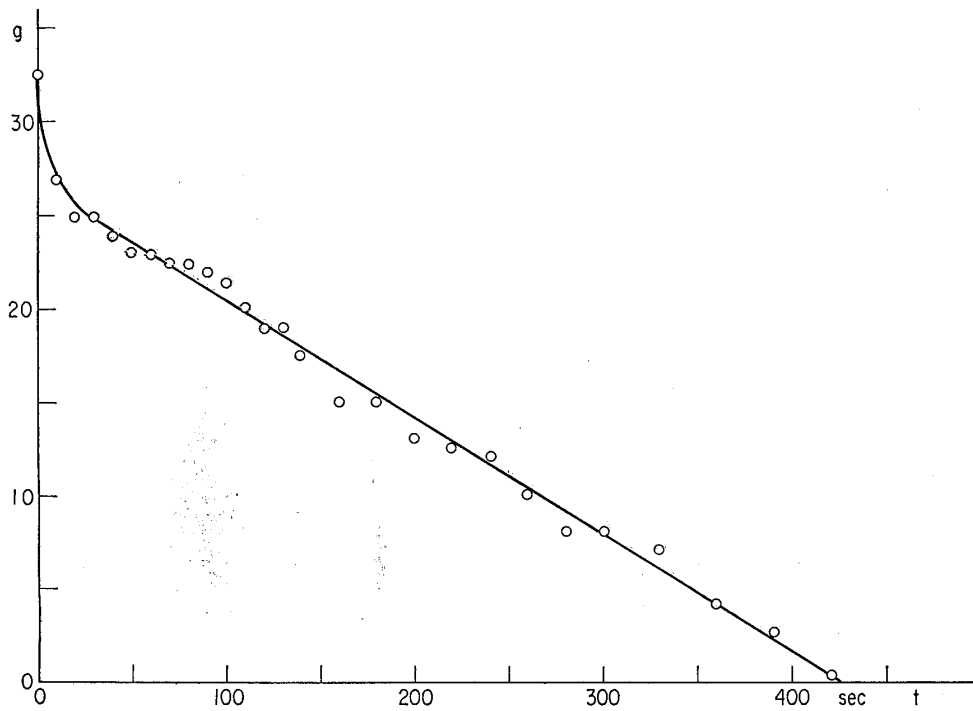


Fig. 5 (A). Time change of total weight (liquid methane alone).

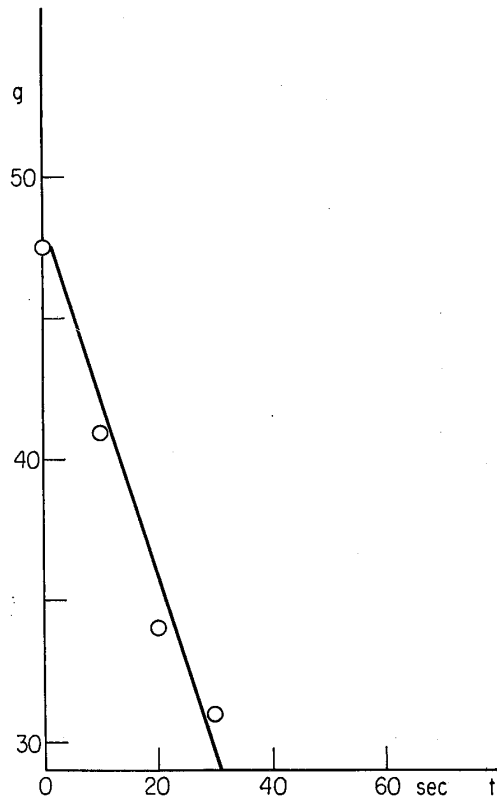


Fig. 5 (B). Time change of total weight (liquid methane poured upon water surface).

insulated vessel by a spring balance.

The result for methane poured directly into the vessel (Case A) is shown in Fig. 5 (A) and that for methane (59 cc) poured upon water (29 cc) (Case B) is shown in Fig. 5 (B). The wind in these experiments were 2 m/sec. ca.

The resulted rates of vaporization were 0.0011 g/sec. cm² for case A, and 0.024 g/sec. cm² for case B. The rate of vaporization in latter case is much larger than in the former case, because the heat supply was much larger than in the former case.

Acknowledgement

We are sincerely grateful to Messrs. S. Nishimura and Y. Tsuru, General Laboratory, Tokyo Gas Co., for their kindness to prepare and let us to make use of liquid methane.

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