

Radioactivity and Radiolysis of the Solid Methane
used as a Cold Neutron Moderator in the KENS Tar-
get Assembly

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Abstract

The radioactivity and radiolysis of the solid methane used as a cold neutron moderator in the KENS target assembly were studied extensively. H_2 , C_2H_4 and C_2H_6 were identified as radiation decomposition products using a gaschromatograph and their concentrations were less than 1 % for the bombardment of 500 MeV protons on the W target at an average beam intensity of 1.2 μA for 52.7 hrs. Only 3H and 7Be were detected as radioactive nuclides in the irradiated CH_4 gas. The concentrations were of the order of $10^{-10} \mu Ci/cm^3$ for 7Be and $8.5 \times 10^{-6} \mu Ci/cm^3$ for 3H . About 60 % of 3H atoms produced was in the chemical form of 3HH and the rest 40 % in tritiated - CH_4 , C_2H_4 and C_2H_6 . Further the 3H concentrations in the cooling waters for the target assembly and the magnets for beam transport were $0.73 \times 10^{-4} \mu Ci/cm^3$ and $\sim 10^{-6} \mu Ci/cm^3$ respectively.

1. Introduction

The cold neutron moderator installed in the KENS target assembly consists of a solid methane of $13^W \times 5^D \times 15^H$ in size, which is placed just upon the tungsten target¹⁾. Fig. 1 shows the details of target assembly. The moderator is cooled down to 20°K by circulation of He gas using a small cryogenerator. When a cycle of two weeks experiments was finished, the methane is evaporated and transferred to a reservoir tank of 0.5 m³, from which the gas is purged into air through a stack every two weeks.

While the W target is bombarded with 500 MeV protons from the booster synchrotron, the solid methane moderator is exposed to high radiations of secondary particles and photons for a relative long period. The informations on the radiation decomposition and the radioactivities of this methane moderator are therefore ~~are~~ required prior to the use at a full power operation.

Recently a short term test was conducted on July 7 to 25 of 1980 to examine the various performances of the KENS facilities. The radioactivity measurement and chemical analysis of the irradiated methane were performed at the same time. Their experimental results are presented in this paper. In addition, the radioactivities of cooling water for the target assembly and the magnets for beam transport are reported.

2. Radiation decomposition of the solid methane

The W target was bombarded at an average beam intensity

of 1.2 μA for 52.7 hrs in the test run. The solid CH_4 exposed to high radiations was transferred to a reservoir tank of 0.5 m^3 after the run. The sample gases to be assayed was taken from this reservoir tank.

The sample was analysed by gaschromatography under the following conditions;

Column: stainless steel 3 mm diameter, 1.5 m long,

Column packing: molecular seive 5 A, 80 ~ 100 mesh,

Column temperature: temperature programmed at the rate of $10^\circ\text{C}/\text{min}$. from 35°C to 200°C , Carrier gas: N_2 , TCD filament current and temperature: 100 mA, 150°C .

Fig. 2 shows the block diagram of the gaschromatograph used in the present experiment.

The retention times of H_2 , CH_4 , C_2H_4 and C_2H_6 gases and their sensitivities to TCD were determined in advance using the standard gases, which were prepared by mixing these gases at various ratios of concentration with a vacuum line. Fig. 3 is the gas chromatogram obtained for the above sample. H_2 , C_2H_4 and C_2H_6 were identified as radiation decomposed products, and their relative concentrations were calculated from the peak area in the gaschromatograms and the sensitivity of each gas to TCD, being presented in Table 1 ($\text{CH}_4=100\%$).

The major product was H_2 , however its concentration was of small quantity less than 1 %. The production ratios of C_2H_4 and C_2H_6 to H_2 in the gas phase radiolysis of CH_4 ²⁾ were lower than the present results. This is indicative that the reactions concerning radicals and/or excited species

containing C atoms take place in the spur more frequently in the solid.

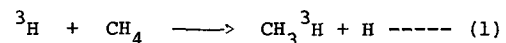
3. Radioactivity of the solid methane moderator

The high energy secondary particles from the W-target interacts with C atoms of the moderator to give spallation reaction products of ^3H (half life: 12.3y) and ^7Be (53 days).

The sample gas taken from a reservoir tank was introduced to a CuO-Oxidation furnace as shown in Fig. 4, and all hydrogen atoms were oxidized and converted to H_2O . The tritiated H_2O collected by a Dry Ice-ethanal trap was measured with a liquid scintillation counter. 2ml of the above water was mixed with 13ml of a Insta-Gel scintillation cocktail, and ^3H -radioactivity was determined by an external standard method. The chemical forms of tritium atoms in the CH_4 moderator were also examined with the apparatus shown in Fig. 4. Tritiated compounds are divided into two fractions of H_2 and organic one (CH_2 , C_2H_4 and C_2H_6) by the use of the liquid N_2 trap placed between the sample gas container and the CuO-oxidation furnace.

The results were presented in Table 2. The ^3H activity in the CH_4 gas reservoir was $8.5 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$ on an average and 40 % of ^3H atoms was in the chemical form of H_2 and the rest 60 % in CH_4 , C_2H_4 and C_2H_6 . The tritium atom produced by spallation reactions has very high translational energy and loses gradually its energy in collisions with surroundings. These hot tritium atom enter the chemical combinations in the energy region less than a few tens of electron volts.

The main reactions of ^3H with CH_4 are H-substitution (1) and H-abstraction (2) reactions³⁾



On the other hand, C_2H_4 and C_2H_6 are considered to be formed in the hot zone, in which a large amount of energy are dissipated and the reactions concerning radicals and excited species take place. The ^3H concentration of the CH_4 gas was greater than the $(\text{MPC})_a$ for ^3H ($2 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$). However the concentrations at the boundaries of site and controlled area can be kept less than the values of KEK radiation rule, $2 \times 10^{-4} \mu\text{Ci}/\text{cm}^3$ and $2 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$ at each position, by releasing the gas through the stack of 20 m in height.

Whereas ^7Be by the reaction of ^{12}C (n, spallation) ^7Be ($E_{\text{th}} \approx 30 \text{ MeV}$, $\sigma \approx 10 \text{ mb}$) is expected to be produced in the same extent to the ^3H activity. However its concentration in the CH_4 gas was of the order of $10^{-10} \mu\text{Ci}/\text{cm}^3$, being extremely small compared with the ^3H activity of $8.5 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$. This indicates that the most ^7Be reacts with CH_4 and/or O_2 present as an impurity to form non-volatile compounds and are adsorbed on the walls of the container and pipes.

The ^3H concentration in the cooling water of the target assembly and the magnets for beam transport were further examined. In order to eliminate ^7Be and other radioactivities, the sample was distilled, and then the water collected at the trap was measured with a liquid scintillation counter in a similar manner as above mentioned. Their concentrations

were $0.73 \times 10^{-4} \mu\text{Ci}/\text{cm}^3$ for the target cooling water and $2 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$ for the magnet cooling water respectively.

References

- 1) "KENS Report-I" ed. by Y. Ishikawa, P.32, (1980).
- 2) K. Yang and P.J. Manno, J. Am. Chem. Soc., 81, 3507 (1959).
- 3) E.A. El-Sayed, P.J. Estrup and R. Wolfgang, J. Phys. Chem., 62, 1356 (1958).

Table 1. Product yields due to the radiation decomposition of the solid CH_4 ($\text{CH}_4=100\%$)

Sample No.	H_2	CH_4	$\text{CH}_2=\text{CH}_2$	CH_3CH_3	O_2
1	0.13 %	100 %	0.03 %	0.05 %	<0.005
2	0.15	100	—	—	<0.003
3	0.15	100	0.04	0.07	<0.003

Table 2 Radioactivity of tritium and chemical forms of tritiated compounds

A) Radioactivity of Tritium

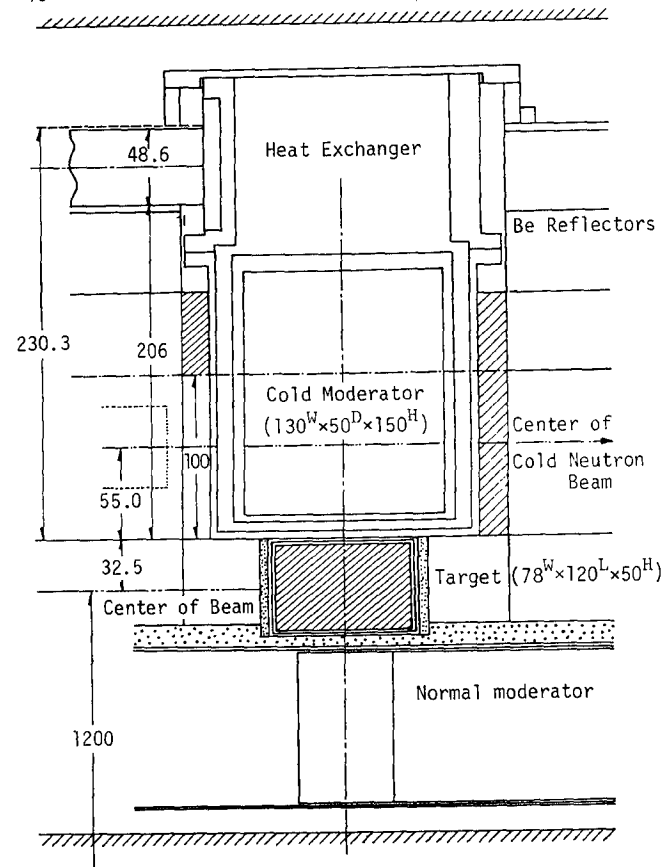
Sample No.	concentration ($\mu\text{Ci}/\text{cm}^3$)
1	8.7×10^{-6}
2	7.9×10^{-6}
3	8.9×10^{-6}
av. $8.5 \times 10^{-6} \mu\text{Ci}/\text{cm}^3$	

B) Chemical Forms of Tritiated Compounds

sample No.	H ₂ -fraction	organic fraction*
1	60.8 %	39.2 %
2	60.9 %	39.1 %
3	59.7 %	40.3 %

* including CH₄, C₂H₄ and C₂H₆

Fig. 1 Side View of KENS Cold Neutron System



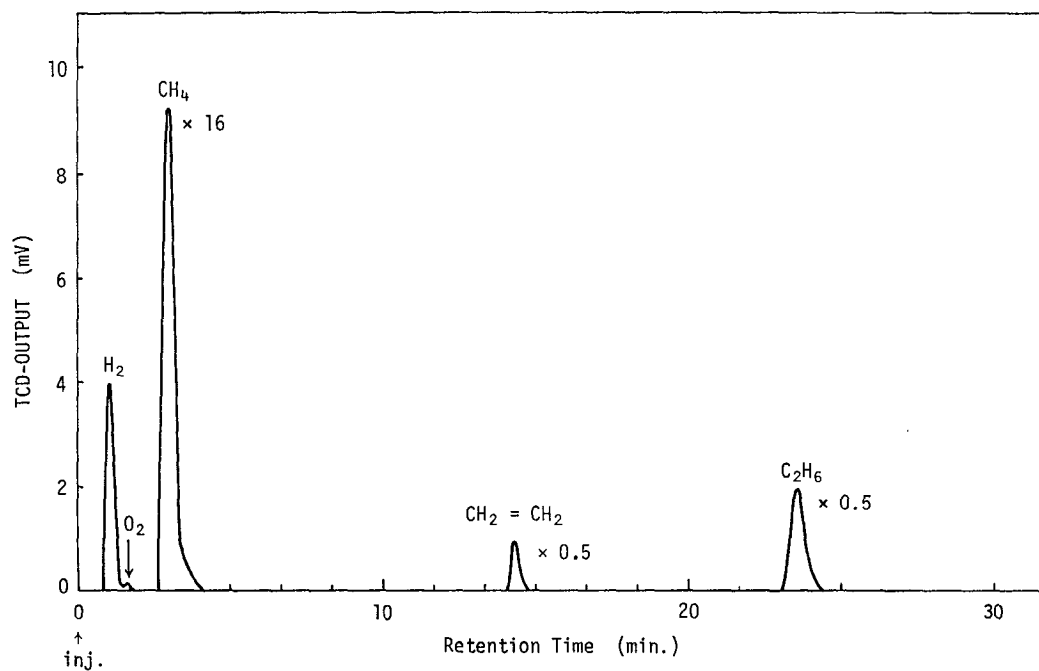


Fig.3 Gas-chromatogram of the irradiated CH₄ moderator

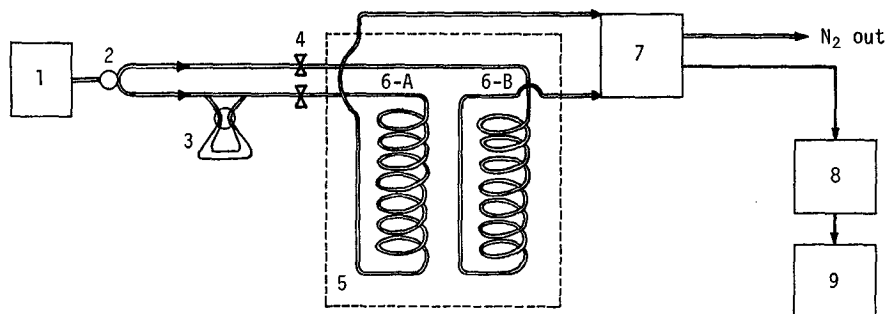


Fig.2 Block diagram of the gaschromatograph

1. N₂ gas container (carrier gas)
2. three-way stopcock
3. sample gas container (10ml) with a four-way stopcock
4. carrier gas flow regulator
5. oven (temperature programmed)
6. 6-A : sample column, 6-B : reference column
7. TCD (thermal conductive detector)
8. amplifier
9. recorder

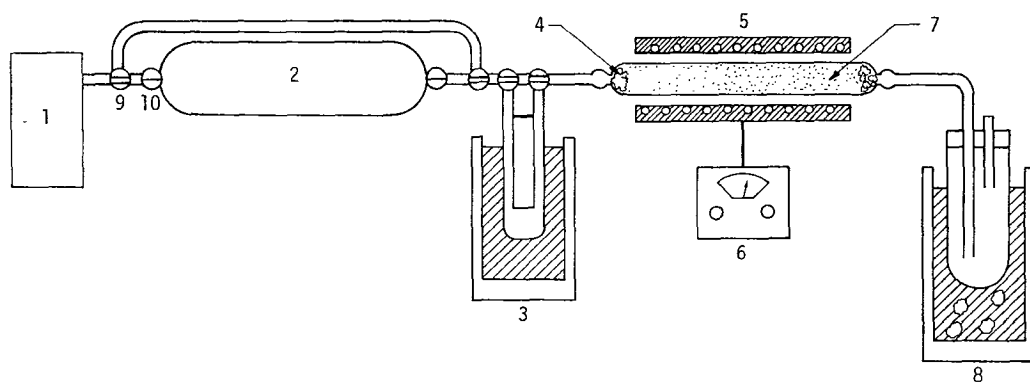


Fig.4 Oxidation apparatus of the sample gas

1. He gas
2. sample gas container
3. liq. N₂ trap
4. glass wool
5. electric furnace
6. temperature controller
7. copper oxide
8. Dry Ice ethanol trap
9. three-way stopcock
10. straight stopcock