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HIGH SENSITIVITY DOSIMETRY BY $^{14}CO_2$

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The gamma dose rate in a nuclear reactor has been measured by using $^{14}CO_2$ as the gaseous chemical dosimeter. High sensitivity of the dosimetry was attained by measurements of the specific radioactivity of the radiolyzed product, ^{14}CO relative to that of $^{14}CO_2$.

Carbon dioxide has been used as the gaseous chemical dosimeter for gamma dose rate measurements in the nuclear reactor[1]. The dose convenient for the volumetry and mass spectrometry of the radiolyzed product, carbon monoxide is several Mrads at least, which dose is not easily attainable in such cases as when the dose rates are not high. In this paper, a new technique was applied for higher sensitivity of the dosimetry, using $^{14}CO_2$.

In a vacuum line, 0.38 ml of $^{14}CO_2$ (specific activity 52 or 59.3 mCi/m mole, The Radiochemical Centre, Amersham), 19.7 ml of unlabeled CO_2 (Research Grade, Takachiho Chem. Ind.) and 0.20 ml of NO_2 (Research Grade, Matheson) were introduced into a quartz ampoule at 1 atmospheric pressure, after trap-to-trap distillation of each component. The ampoule was irradiated in the "Grazing Hole" of the nuclear reactor "Yayoi" at 500 W for 1 or 2 h or at 1 kW for 2 h. The gas in the ampoule was sampled in a vacuum line and injected into a radiogaschromatograph (He carrier gas (20 ml/min), Porapak Q column 4 mm i.d. and 3 m long. C_3H_8 (20 ml/min) added to the He stream after separation before a gas-flow counter, 23 mm i.d. and 30 cm long, operated at 4400 V). Retention times of ^{14}CO and $^{14}CO_2$ were 4.5 and 15 min and the separation of the peaks was complete.

The ratio of the radiogaschromatograph peak areas for ^{14}CO and $^{14}CO_2$ gave the decomposition ratio (DR, hereafter) of the irradiated gas. The control samples showed definite DR, depending on the interval between the sample preparation when the back reaction inhibitor NO_2[2] was added to CO_2 and analysis due to the β radiolysis by ^{14}C. By subtracting the time proportional contribution of the self irradiation effect from the observed DR, the absorbed dose, D(Mrad) of the CO_2 dosimeter could be calculated by an equation; $DR = 2.28 \times 10^{-4} D$, assuming the G(CO) value to be 5.0[1], [3]. The experimental results and the obtained gamma dose rates are shown in the Table, from which the linearity of the CO_2 decomposition to the exposure is seen and the sensitivity and accuracy of the present method can be estimated.

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Table. Decomposition Ratios of Irradiated Samples

No	Interval(h)	$^{14}CO/^{14}CO_2$ Ratio (%) Obs.	Average*1	Irradiation Power*2	Time(h)	Dose Rate (Mrad/h)*3
1	27.0	0.0217	0.0133	500 W	1	1.17
2	41.3	0.0387				
	43.3	0.0414	0.0270	500 W	2	1.18
3	45.2	0.0689				
	47.2	0.0672	0.0538	1 kW	2	1.16

*1 Corrected for decomposition by self irradiation (correction factor 0.00031 %/h) and averaged.

*2 Reactor thermal power.

*3 Normalized for a reactor power of 1 kW.

In the radiogaschromatograph, a sharp shoulder peak (retention time, 10.5 min) appeared for all samples, which areas corresponded to several percents of the main ($^{14}CO_2$) peak areas. Mass spectrometry of the sample showed no peak in the m/e range between 68 (= $^{12}C_3O_2$) and 74 (= $^{14}C_3O_2$). Also, conventional TCD gaschromatography with several kinds of columns could not explain the unidentified peak in the radiogaschromatograph.

In the present experiment, 1 mCi of $^{14}CO_2$ was sealed into an ampoule, diluted about 50 times with unlabeled CO_2 and about 1/20 of the irradiated gas was sampled for an analysis. By absorption of 1 Mrad, 1 mCi of $^{14}CO_2$ forms ^{14}CO with an activity, 8×10^3 dps. An amount of ^{14}CO equivalent to about 400 dps was used for an analysis, which was about 20 times larger than the background counting rate of the gas flow counter employed. In this sense, a dose of about 50 krad can be detected with the present method, and a lower dose with larger sampling ratios can be expected. On the other hand, there is an inherent problem of self irradiation as a background in the measurement of external irradiation dose. Dilution with unlabeled CO_2 reduces the self irradiation dose rate, which can be about 200krad/h, if 100 % $^{14}CO_2$ isotope concentration, 100 % absorption of ^{14}C β rays (0.155 MeV), 1/3 effective energy of the β energy and G(CO) = 5.0 are to be assumed. But the dilution has an adverse effect of lowering the specific activity in the dosimetric mixture. Thus, an appropriate dilution ratio and analytical conditions (volumes to be irradiated and sampled, counter sensitivity and the time interval between sample preparation and analysis) should be chosen for the highest sensitivity, which would be several tens of krads and 1 to 2 orders of magnitude higher than that of the conventional non radioactive CO_2 dosimetry.

References

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