DETERMINING CARBON-13 ENRICHMENT USING LOW ENERGY PROTONS*

Zhang Weicheng (张维成), Chao Zhiyuan (晁致远) and Peng Xiuru (彭秀茹)
(*Institute of Modern Physics, the Chinese Academy of Sciences, Lanzhou 730000, China*)

ABSTRACT

The proton-capture reactions $^{12}C(p,\gamma)^{13}N$ and $^{13}C(p,\gamma)^{14}N$ have been studied to determine ^{13}C enrichments. The system has been calibrated by measuring the gamma-rays yield from the $^{12}C(p,\gamma)^{13}N$ and $^{13}C(p,\gamma)^{14}N$ reactions as a function of known ^{13}C enrichment. This technique is applicable to the analysis of samples with ^{13}C enrichments between 1% and 90%.

Keywords Proton-capture reactions, Carbon-13, Isotope ratio, Gamma detection

1 INTRODUCTION

The use of carbon tracer studies is well established, particularly as applied to biological systems. Much of this experience has been gained through the use of the radioactive isotope ^{14}C. In fact, as stable isotopes have become more advantageous and more extensive, recent efforts have results in a dramatic increase in the availability of the stable isotope of carbon [1~4]. Methods for detecting the radioactive tracers are extremely sensitive. If suitable detection schemes can be developed for the stable isotopes, their application is assured. There has, therefore, been considerable effort recently in developing many such techniques[5,6]. It is to this area that we have devoted. The aim of this work is determination of ^{13}C enrichment using low energy protons.

2 EXPERIMENTAL

The experiment was performed using the 2×1.7MV tandem accelerator at a proton energy of 600 keV and beam currents of about 0.5 mA. The barium carbonate targets of known ^{13}C enrichments determined by mass spectrometry were bombarded. The various ^{13}C enrichments of $BaCO_3$ used are listed in Table 1. The barium carbonate was pressed directly into 2 mm thick disks of 12 mm diameter, and tightly mounted onto the target holder which was positioned at zero degree relative beam axis. The target holder allows up to ten samples to be analysed. Several cross reference normalization measurement were made using natural graphite. The pulses from the detector were fed to IBM PC data acquisition system and the spectra stored in the memory for analysis.

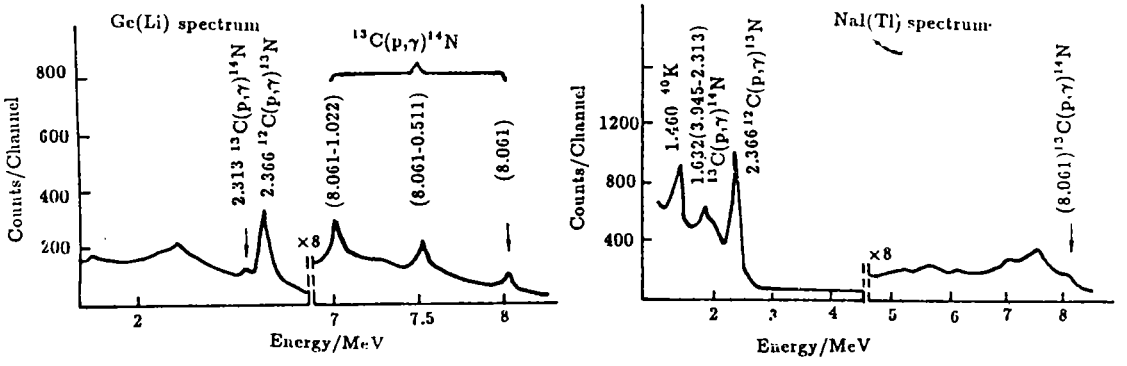
Table 1
Targets of enriched $BaCO_3$

Target	^{13}C (wt) /%	Target	^{13}C (wt) /%
$BaCO_3$	0.7	$BaCO_3$	73.00
$BaCO_3$	10.0	$BaCO_3$	85.00
$BaCO_3$	26.57	Graphite	1.11
$BaCO_3$	58.45		

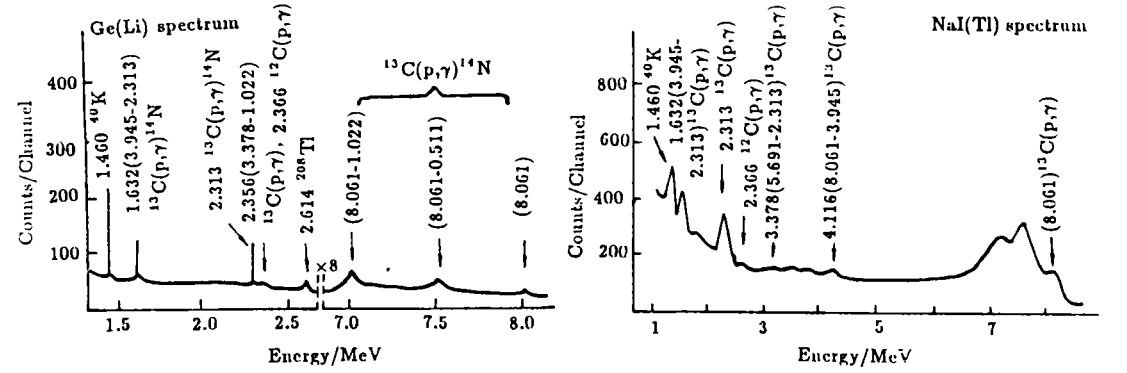
*The Project Supported by National Natural Science Foundation of China
Manuscript received date: 1995-01-10

3 RESULTS AND DISCUSSION

Typical Ge(Li) and NaI(Tl) data are shown in Figs.1~4 for a natural abundance target and for a target highly enriched with ^{13}C, respectively. It is seen by comparing the spectra in Figs.1, 2 that the "^{12}C peak" in NaI(Tl) spectrum is composed predominantly



Figs.1,2 Typical Ge(Li) and NaI(Tl) spectra for 600 keV protons incident on infinitely, natural graphite carbon target



Figs.3,4 Typical Ge(Li) and NaI(Tl) spectra for 600 keV protons incident on infinitely, highly enriched carbon target

of 2.366 MeV gamma-rays from the $^{12}C(p,\gamma)^{13}N$ reaction. For higher ^{13}C enrichments, as in Figs.3, 4, the "^{12}C peak" in the NaI(Tl) spectrum is composed almost entirely of the 2.313 MeV gamma-rays from the $^{13}C(p,\gamma)^{14}N$ reaction, with the 2.370 MeV and the double escape peak of the 3.378 MeV gamma-rays also making significant contributions to the ^{12}C yield. In the Ge(Li) spectra, prominent full energy, single escape, and double escape peaks are seen,, and these assays made more accurately. On the other hand, because of nature and size of the detector, these spectra in the NaI(Tl) spectra show only a strong full energy and single escape peak, the double escape peak appears as a broadening on the low energy side of the single escape peak. It is noted the change in the vertical scale for these spectra, and implying that the use of a large NaI(Tl) detector is necessary when large batches of samples have to be examined quickly.

Table 2 shows that the lower resonances in both ^{12}C and ^{13}C are particularly attractive for determining carbon isotopic abundance. The 457 keV resonance in ^{12}C populates the level in ^{13}N at 2.366 MeV. The 554 keV resonance in ^{13}C populates the 8.061 MeV excited state in ^{14}N. The decay-of $^{13}N^*$ result is only the 2.366 MeV gamma-rays. The decay-of $^{14}N^*$ results are not only the 8.061 MeV gamma-rays, but also a 2.313 MeV gamma-rays, a 2.370 MeV gamma-rays, and a 3.378 MeV gamma-rays which has a double escape peak at 2.356MeV. Fig.5 shows the 2.3MeV region of a Ge(Li) spectrum for 600 MeV protons

Table 2

Main characteristics of nuclear reaction $^{12,13}C(p, \gamma)^{13,14}N$

Reactions	Thresh. /MeV	Proton energy /keV	Half-widths /keV	Max cross-section /mb	Prominent gamma-rays/MeV
$^{12}C(p,\gamma)$	1.944	456.8±0.5, 1698±5	39.5±1, 72±9	0.127, 0.035	2.366, 3.509
$^{13}C(p,\gamma)$	7.550	554±1	32.5±1	1.44	8.061,4.116,3.378,2.370,2.313,1.632
		1747.6±0.9	0.075±0.050	340	9.172,7.028,6.444,2.728,2.144

incident on an infinitely thick, highly enriched (85% ^{13}C) carbon target. The 2.366 MeV gamma-rays are from the $^{12}C(p,\gamma)^{13}N$ reaction, the other gamma-rays are from the $^{13}C(p,\gamma)^{14}N$ reaction. The "FE" stands for a full energy peak and "P-2" stands for a double escape peak. These 2.3 MeV gamma-rays from $^{14}N^*$ interfere with the 2.366 MeV gamma-rays from $^{13}N^*$. If a high resolution Ge(Li) detector is used as the gamma-rays spectrometer, 2.313 MeV gamma rays from $^{14}N^*$ can be resolved from the 2.366 MeV gamma-rays from $^{13}N^*$. However, when a NaI(Tl) detector is used, these two rays cannot be resolved.

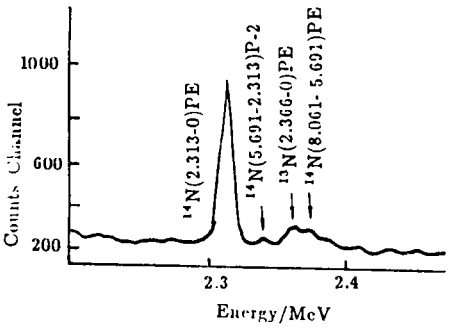


Fig.5 The 2.3 MeV region of a Ge(Li) spectrum

As can be seen from Table 2, the width of the ^{12}C resonance at E_p=457 keV is 39 keV. Thus, the line associated with the 2.366 MeV gamma-rays has a natural width of 39 keV. Consequently, this gamma-rays cannot be resolved from the 2.370 MeV gamma-rays and the double escape 2.356 MeV gamma-rays resulting from the decay of $^{14}N^*$. The 2.370 MeV gamma-rsys result from a transition from the resonance state at 8.061 MeV to an excited state at 5.691 MeV. The ^{13}C resonance at E_p=554 keV has a natural width of 32 keV, so the line associated with the 2.370 MeV gamma-rays has a natural width of 32 keV. However, the 2.356 MeV double escape gamma-rays, having a full energy of 3.378 MeV state, result from a transition between the 5.691 MeV state and 2.313 MeV state. Since these are bound states, the line shape associated with this gamma-ray transition is sharp, the observed line width being determined by the detector resolution. All of the gamma-rays discussed are labeled, Fig.5 shows explicitly the difficulty in stripping out the peaks in 2.3 MeV area. If NaI(Tl) detectors with its poorer resolution were used, none of the peaks in Fig.5 would be resolved.

Thus, when one attempts to measure the abundance of carbon in a target. The carbon-13 is determined very simply and reliably using the 8.061 MeV gamma-rays due to the $^{13}C(p,\gamma)^{14}N$ reaction does produce a clean line with no interference. It is, therefore, convenient to use the area under the peak associated with this gamma-rays as a measure of the amount of ^{13}C present. For the carbon-12, the experimenter does not get a pure ^{12}C signal since the characteristic 2.366 MeV line is not isolated in the gamma-rays spectrum of a carbon target bombarded with 600 keV protons, certain gamma-rays from $^{14}N^*$ interfere with the 2.366 MeV gamma rays from $^{13}N^*$. These gamma rays as pointed out above, being in reality a complex mixture of several lines. Figs.6,7 give calibration

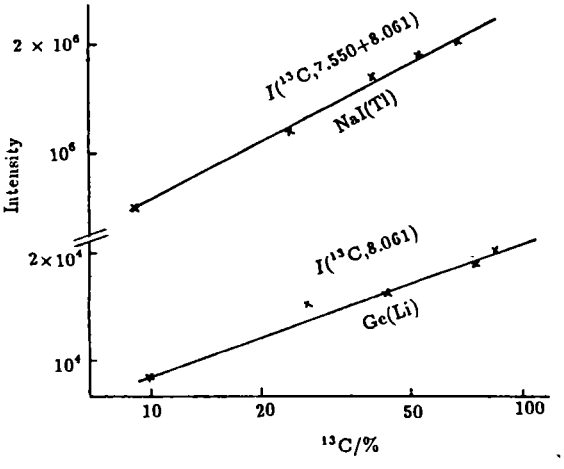


Fig.6 Intensity of characteristic ^{13}C line vs its abundance in $BaCO_3$
E_p=600 keV I =0.5 mA

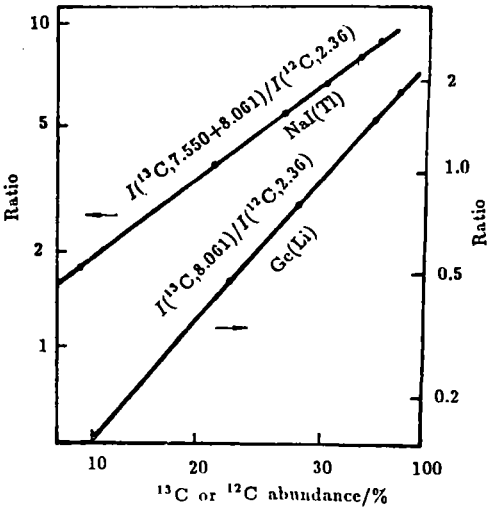


Fig.7 Characteristic line intensity ratio of ^{13}C to ^{12}C vs ^{13}C abundance in $BaCO_3$, E_p=600 keV I =0.5 mA

curves established with NaI(Tl) and Ge(Li) detectors. These curves show clearly that although the Ge(Li) detector, as expected, is apparently the more suitable for such analysis, the NaI(Tl) detector can be useful for some isotopic measurements, especially at carbon-13 contents below 70%. A point to emphasize in this respect is that while the former instrument is more selective because of its resolution. The latter, owing to its efficiency, reduces the measurement time by a factor 10 to 20. This is important advantage when large batches of samples must be examined quickly at costs competitive enough to warrant the use of these techniques in place of mass spectrometry.

REFERENCES

1 Klein P D, Klein E R. Stable isotopes. Amsterdam:Elsevier, 1982; 347
2 Schoeller D A, Scheneider J F, Solomons N W *et al.* J Lab Clin Med, 1977; 90:412
3 Xia Zongqin, Dai Tengchang, Hu Ya'er *et al.* Nuclear Techniques (China). 1985; 3:1
4 Yang Zhizhong, Zhu Deping, Lin Chun *et al.* Chi J Nucl Med, 1990; 10(4):23
5 Zhang Weicheng, Lei Xiangguo. Int J Radiat, Appl Instr, 1991; A42(11):1039
6 Zhang Weicheng, Zhang Tianei, Lei Xiangguo. J Radioanal Nucl Chem Artic, 1991; 155(2):313