DETERMINATION OF ^{13}C IN BREATH SAMPLE BY PIGE*

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ABSTRACT

This paper discusses a novel application of PIGE for the determination of ^{13}C in breath sample. Samples of human breath, urea, glucose, benzamide, barium carbonate were analyzed against tank CO_2 and graphite standard. An accuracy check of the ^{13}C determination (with reference to mass spectrometric "True" results) gave a relative error of only- 0.4% for PIGE. The performance of different standard in this determination was assessed. Relative standard deviation for the determination of ^{13}C isotopic abundance in breath samples were <20%. Then, if a 25% change is conservatively assumed observable in ^{13}C abundance, an increase in ^{13}C percent isotopic abundance from the natural 1.11% (average) to only 1.39% may be detected.

Keywords: PIGE Breath sample

I. INTRODUCTION

The proton induced gamma- ray emission (PIGE) methods is suitable for analysis of light elements using a low energy accelerator[1]. The purpose of this paper is to report the feasibility and the applicability of PIGE as a conventional method offered by the nuclear reaction $^{12}C(p,\gamma)^{13}N$ and $^{13}C(p,\gamma)^{14}N$ to determine ^{13}C in breath samples when using ^{13}C as a tracer in breath test. ^{13}C is nonradioactive, its advantage over ^{14}C is clear.

II. EXPERIMENTAL

We bombarded thick samples of human breath, urea, glucose, benzamide, barium carbonate, as well as thick graphite and tank CO_2 standards, with 0.6MeV proton from a 2× 1.7 or a 2× 2 Tandem. Prompt gamma- ray spectra produced during nuclear reactions with ^{12}C, ^{13}C were measured with NaI(Tl)- IBM- PC data acquisition system [2]. The intensities of the photopeaks of interest were calculated in each of the spectra with the help of the program MMCA- CM8 at IBM- PC- XT computer. The detector was placed immediately behind the target so as to provide a maximum solid angle. Reactions used, their corresponding resonance energies and prompt γ - ray are $^{12}C(p,\gamma)^{13}N$, 0.457MeV, 2.366MeV; $^{13}C(p,\gamma)^{14}N$, 0.554MeV, 8.061MeV, respectively[3]. As only thick targets were used, the 0.6MeV protons could only reach (by energy degradation in them) the 457keV and 554keV resonances of the reactions $^{12}C(p,\gamma)^{13}N$ and $^{13}C(p,\gamma)^{14}N$ respectively; thus, the intensities of the corresponding 2.366MeV and 8.061MeV gamma ray were correlated with the contents of ^{12}C and ^{13}C, respectively, in samples.

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Ⅲ. RESULTS

1) The background ascertainment The background was measured both when the accelerator is off and with 0.5μ A beams of 0.7– 1.5 MeV protons on blank copper and quartz targets. No significant variations were observed, so that the background was attributed to the accelerator building rather than to the machine operation. The 1.46 and 2.61 MeV Gamma– ray, respectively, from ^{40}K and ^{208}Tl (^{232}Th decay chain) in the concrete walls, were always present. Background spectra to be subtracted were always counted shortly before or after the corresponding sample or standard spectra.

2) Yields and sensitivities The natural graphite were irradiated at various proton energies to determine Gamma ray yields for the reactions of interest. The results obtained are presented in Table 1. Table 1 shows little energy dependence of the gamma– ray yields for ^{12}C and ^{13}C. During elevating bombarding energy, the background under the ^{12}C peak increase, and ^{13}C peaks become less clearly defined and inconveniently close to interfering radiation. Therefore bombardment with 0.6 MeV protons yields satisfactory sensitivities for ^{12}C and ^{13}C, without suffering significant interference.

Table 1

Gamma–ray yields from the reaction ^{12}C (p,γ) ^{13}N ^{13}C(p,γ) ^{14}N and Related analytical parameters

Determination	Units	Reacting nuclide	Proton bombarding energy (MeV) 0.6	0.7	0.8	0.9
Yield	cpm/μ A	^{12}C	5460.8	4559.6	3126.1	4094.21
		^{13}C	1009.4	1070.5	1074.4	1069.2
Sensitivity	cpm/ (μ Appt*)	^{12}C	5.52	4.61	3.16	4.14
		^{13}C	91.1	96.6	96.9	96.5
Detection limit* *	ppt	^{12}C	3.69	4.79	6.09	4.69
		^{13}C	0.109	0.113	0.112	0.110

* 1 ppt = 1 part per thousand * * Detection limit = (Minimum determinable activity per μ A)/(sensitivity). Assume minimum determinable activity given by 3 times the square root of the background in the energy window of interest.

3) Accuracy and nature of the standard Classical studies on differences in ^{13}C isotopic abundances amoung common natural substances indicate a range of variation of 4.5%[4]. Therefore, the accuracy of the present method has to be checked against previous mass spectrometry (MS) determinations, taken as "true" results in this work. The mean ^{13}C per cent abundance was obtained for $BaCO_3$, by PIGE, with reference to a "true" graphite standard, and compared with a "true" results obtained by MS for the same $BaCO_3$ sample. The result is presented in Table 2. The relative deviation was only – 0.4% in this test.

The accuracy depends also on the nature of samples and standard. In general, the best results may be obtained when standard and sample have similar stopping power characteristics. A comparison of all the substances used in this study is shown in

Table 2

The comparison of ^{13}C abundance determined by MS and PIGE

Method	Reference standard values	^{13}C abundance in $BaCO_3$
MS(MAT- 251)	1.108	1.084
PIGE	1.108	1.089

Table 3, whose figures are percent deviations between the proton induced gamma- ray yield ratio $Y(^{13}C)/Y(^{12}C)$ for a given substance (R_o) and the analogous ratio (R) for each of others; i.e., 100 $(R_o- R)/R_o$ in each comparison. It may be assumed that ^{13}C abundance variation among biological and organic substances is small; thus, the small deviations between each other may be attributed to similar stopping- power effects[5]. In other words, a known biological tissue can be an effective standard for an unknown of analogous nature.

Table 3

Comparison between carbon isotopic yield ratios for several substances

Substance	Benzamide	Urea	Graphite	Tank CO_2	Glucose	Human breath (Woman)	Human breath (Man)
Benzamide	0	- 9.7	- 9.7	- 1.7	- 4.3	- 1.6	- 5.9
Urea	+8.8	0	- 0.8	+7.3	+4.9	- 5.7	+3.4
Graphite	+8.8	+0.8	0	7.3	+4.9	- 5.6	+0.4
Tank Co_2	+1.7	- 7.8	- 7.9	0	- 2.5	- 14.0	- 4.2
Glucose	+4.1	- 5.1	- 5.2	+2.5	0	- 11.1	- 1.5
Human Breath (Woman)	+13.7	+5.4	+5.3	+12.2	+9.9	0	+8.6
Human Breath (Man)	+5.5	- 3.5	- 3.6	+4.0	+1.5	- 9.5	0

4) Determination of ^{13}C in breath samples ^{13}C abundance of human breath natural samples was determined by comparison with the two standards. Each sample was bombarded three successive times with about a 0.5μ A beam of 0.6 MeV proton. Fig.1 and 2 show typical spectra for standard and sample. The sample spectra resemble each other and also similar to the standard. In all spectra the peaks of interest are well defined and free from interferences, thus being quite suitable for countable comparisons. Some results are presented in Table 4.

The standard deviation indicated in Table 4 are taken to represent the precision of these determination. It may be observed that the precision values for breath samples is $<20\%$ and these precision values are independent of the nature of the standard used, while the absolute error (accuracy) is generally below these value, as expected. Then, if a 25% change is conservatively assumed observable in ^{13}C abundance, an increase from its natural 1.11% value to 1.39% will be detected.

Table 4

Precision and accuracy of ^{13}C abundance determinations in human breath samples by 0.6 MeV proton

Sample	Standard	Average ^{13}C abundance (%)	Standard deviation (%)	Absolute error* (%)
Human breath (woman)	Graphite	1.17±0.23	19.5	+5.6
	Tank CO_2	1.18±0.23	19.6	+6.5
Human breath (man)	Graphite	1.04±0.13	12.4	- 6.1
	Tank CO_2	1.01±0.15	14.7	- 8.8

* All samples had the natural abundance of ^{13}C, that is, 1.108%

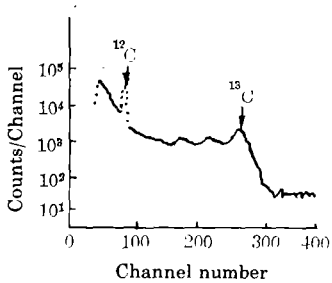

Fig.1 Typical prompt γ -ray spectra of graphite under 0.6 MeV proton bombardment

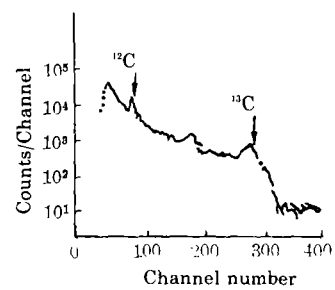

Fig.2 Typical prompt γ -ray spectra of human breath samples under 0.6 MeV proton bombardment

The isotopic abundance for ^{13}C in breath samples were the natural values and, thus, the minimum to be possibly encountered in most stable tracers experiments; they can only increase by administrating stable tracers to living organisms, with corresponding improvements in counting statistics and, therefore, presision in actual practice.

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