# MEASUREMENT OF 018/016 RATIO USING A FAST NEUTRON REACTOR

H. G. Jackson, et al

R and D Associates

Prepared for:

Advanced Research Projects Agency

February 1973

R & D Associates
P.O. Box 3580
Santa Monica, California
90403

RDA-TR-136-NMO



# MEASUREMENT OF $O^{18}/O^{16}$ RATIO USING A FAST NEUTRON REACTOR

by

H. G. Jackson, L. M. Libby, and H. R. Lukens

February 1973



Sponsored by
Advanced Research Projects Agency
ARPA Order No. 1964-1

ARPA Order No. - 1964-1
Program Code - 3F10
Name of Contractor - R & D Associates
Effective Date of Contract - 1 November 1972
Contract Expiration Date - 31 October 1973
Amount of Contract Dollars - $99,975.00
Contract Number - F44620-73-C-0029
Principal Investigator - Leona Libby
Phone Number - 213-451-5838
Program Manager - Leona Libby
Phone Number - 213-451-5838
Short Title of Work - Historical Climatology



This research was supported by the Advanced Research Projects Agency of the Department of Defense and was monitored by the Air Force Office of Scientific Research under Contract No. F44620-73-C-0029.

Security Classification

DOCUMENT CONTROL DATA - R & D

*(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)*

| ORIGINATING ACTIVITY (Corporate author) | 2a. REPORT SECURITY CLASSIFICATION |
|---|---|
| R & D Associates | unclassified |
| | 2b. GROUP |

3. REPORT TITLE

"Measurement of $O^{18}/O^{16}$ Ratio Using a Fast Neutron Reactor"

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Semi Annual Technical Report, November - April, 1973

5. AUTHOR(S) (First name, middle initial, last name)

H. G. Jackson, L. M. Libby and H. R. Lukens

| 6. REPORT DATE | 7a. TOTAL NO. OF PAGES | 7b. NO. OF REFS |
|---|---|---|
| February, 1973 | ten | two |

| 8a. CONTRACT OR GRANT NO. | 9a. ORIGINATOR'S REPORT NUMBER(S) |
|---|---|
| F44620-73-C-0029 | RDA-TR-136-NMO |
| b. PROJECT NO. | |
| c. | 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report) |
| d. | AFOSR - TR - 73 - 1000 |

10. DISTRIBUTION STATEMENT

| 11. SUPPLEMENTARY NOTES | 12. SPONSORING MILITARY ACTIVITY |
|---|---|
| | Advanced Research Projects Agency<br>1400 Wilson Boulevard<br>Arlington, Virginia 22209 |

13. ABSTRACT

A method has been devised and proven to measure the stable isotope ratio $O^{18}/O^{16}$, for oxygen, by neutron activation in a fast neutron reactor. The measurement is made non-destructively on a few grams of water with an accuracy of a few parts in ten thousand. At present, measurement to this accuracy requires about 40 minutes with the reactor running at a power of 250 kilowatts, but by increasing detector sensitivity the method promises to become a routine matter accomplished in less time.

DD FORM 1 NOV 65 1473

UNCLASSIFIED

Security Classification

## ABSTRACT

A method has been devised and proven to measure the stable isotope ratio $O^{18}/O^{16}$, for oxygen, by neutron activation in a fast neutron reactor. The measurement is made non-destructively on a few grams of water with an accuracy of a few parts in ten thousand. At present, measurement to this accuracy requires about 40 minutes with the reactor running at a power of 250 kilowatts, but by increasing detector sensitivity the method promises to become a routine matter accomplished in less time.

## INTRODUCTION

If rapid, non-destructive ways of measuring ratios of stable isotopes were available, the use of stable isotopes in research would be greatly aided. We have shown that, for the stable isotopes of oxygen, the ratio $O^{18}/O^{16}$ can be measured non-destructively in small amount. of water, by neutron activation. The activities measured are:

(a) $O^{16}(n,p)N^{16}$; threshold neutron energy = 10.25 MeV,

$\sigma_{16}(n,p)$ = 40 mb, $t_{16}$ (mean life) = 10.3 sec

$E_\gamma$ = 6.14 MeV (68%)

(b) $O^{18}(n,\gamma)O^{19}$; formed by thermal neutron capture,

$\sigma_{18}(\gamma,n)$ = 0.22 mb, $t_{18}$ (mean life) = 41.8 sec

$E_{\gamma 1}$ = 1.37 MeV (60%), $E_{\gamma 2}$ = 200 KeV (100%)

It is necessary to irradiate the water in a neutron reactor which has a high fractional flux of fast neutrons (relative to that available in the graphite-moderated reactors) in order that the 10 second activity has sufficient intensity. Its gamma ray has a high energy, 6.14 MeV, and so is relatively easy to measure, because the background activities from impurities in water and from the Compton radiation are small at this energy. The 42 second activity has two gamma rays, namely 1.37 and 0.2 MeV. That at 1.37 MeV would be the more desireable to measure, except that it coincides with a gamma ray from neutron-activated sodium (which is an abundant impurity) so closely that it can not be identified separately. Therefore this activity was measured by counting the

lower energy gamma radiation at 200 KeV. Here the Compton background is large and is the factor limiting the accuracy of each determination.

The 10 second and the 42 second activities are measured at the same time in the same detector, being separated by pulse height analysis, and recorded separately. Water samples in which it is desired to measure the ratio $O^{18}/O^{16}$ are compared with a standard water sample, and the ratio relative to the standard is expressed as a deviation $\delta^{18}$ in parts per thousand (‰), according to the definition,

$$\delta^{18}(\text{‰}) = \left\{ \left( R_{meas} / R_{standard} \right) - 1 \right\} \times 1000 \qquad \text{(Eqn. 1)}$$

The desired accuracy is obtained by irradiating and counting the induced activities of the water sample several times until some hundred million gamma rays have been counted.

## EXPERIMENTAL

The TRIGA Mark I reactor in steady state operation at 250 kilowatts has a thermal neutron flux of $2.8 \cdot 10^{12}/cm^2 sec$, and a fast (fission spectrum) flux of $3.5 \cdot 10^{12}/cm^2 sec$ at the irradiation position. Water samples of approximately 5 grams each, in plastic vials[1] containing no intrinsic oxygen, were shot into the reactor in the fast pneumatic transfer system and there irradiated for 30 seconds, after which they were shot out into a reproducible site in the counting facility. After an interval of 10 seconds, the induced 10 second and 42 second activities were counted during the next subsequent interval of 30 seconds.

Reactor power is maintained at a preset level by servo-regulated control rod drives. The insertion of 5 grams of water in plastic vials into the outermost fuel element ring (sample irradiation position) is equivalent to adding a slight amount of reactivity to the core, which is immediately compensated for by automatic control rod adjustment without overshoot. The reactor power is maintained to $\pm$0.13% of the preset level at a confidence level of 95%. The temperature at the sample position ranges from 20-25$^{o}$C during the year, but is held constant during any given hour of reactor operation by forced circulation of pool water.

The technology and use of the reactor in similar measurement has been described in Reference 2.

The count in the 200 KeV channel was at a rate of 0.2 Megacycles, and this is too fast for standard pulse height selection equipment. To handle the fast count rate, a simple pulse height analyser was built, using integrated circuitry, according to the block diagram shown in Figure 1. The system is limited to a maximum count rate of 300 MC by the scalers. The sensor was a 1/2" high x 2" diam Na I crystal glued with a high viscosity fluorocarbon (Dow Corning 20-057) to a photo multiplier, RCA-6810 A.

The counts observed in 5 gm of normal water (Colorado River water from the public water system of La Jolla, California, for which $O^{18}/O^{16}$ = 0.2493% as measured in a mass spectrograph ), in a counting interval of 30 seconds, for the 200 KeV radiation (channel 2) and the 6.1 MeV radiation (channel 4), are listed in Table I. Counting ratios measured at two different power levels of the reactor, namely at 190 and 250 KeV, are strictly comparable, showing that counting losses were negligible. Varying amounts of water enriched in $O^{18}$ were added to normal water to prepare a series of samples in which $O^{18}$ was progressively concentrated. The measured ratios of counts in the two channels recording the 200 KeV and the 6.1 MeV radiation respectively are listed in Table II, for

increasing concentrations, and are shown to be linearly dependent on the $O^{18}$ concentration. As shown in Table I. after 24 runs, each of 70 seconds, the $O^{18}/O^{16}$ ratio in a given water sample was measured to an overall error of $\pm$0.04% after 24 determinations, each of standard deviation 0.13%. No external perturbations were allowed to influence the reactor during the measurements. The overall standard deviation of $\pm$.04% testifies to the constancy of the ratio of fast neutron flux.

The enriched water was measured by mass spectrograph[3].

The pneumatic transfer apparatus, TRIGA Mark I reactor, and timing system have been used as unchanged components in a U-235 measurement procedure that delivered 0.37% precision in individual measurements[2]. Hence, the variation in individual channels is believed to be controlled by the counting equipment. Fortunately, the count ratio proved to be largely independent of the individual channel's variance. However, it is likely that improvements in counting equipment stability and counting efficiencies will improve both the speed and precision of the isotopes ratio measurement.

## CONCLUSION

A non-destructive method of measuring the stable isotope ratio $O^{18}/O^{16}$ in water has been shown to be possible, and rapid, and feasible as a commercial service, presently accurate to about 4 parts in 10,000, at a reasonable expenditure of measuring time (about 40 minutes at present), and to promise future measurements at least to this accuracy in shorter times, when the detector sensitivity has been increased.

## TABLE CAPTIONS

Table I Observed decays proportional to concentrations of $O^{18}$ and of $O^{16}$, and their ratios, for 24 experiments on faucet water from the public water supply of La Jolla, California.

Table II Observed decays proportional to concentrations of $O^{18}$ and of $O^{16}$ in water samples sequentially enriched in $O^{18}$, plotted versus $O^{18}/O^{16}$ computed from mass spectroscopic measurements.

## FIGURE CAPTIONS

Fig. 1 Schematic diagram of pulse height selector, and recording scalers, assembled to measure megacycle count rates without significant losses.

TABLE I. Counting Data*, Normal Water

| Date | Reactor Power, KW | Sample Number | Net Weight, Grams | Net Counts Channel 2 | Net Counts Channel 4 | X, Count Ratio Channel 2/Channel 4 |
|---|---|---|---|---|---|---|
| Sept. 7 | 190 | 1 | 4.9640 | 3,793,063 | 531,946 | 7.1305 |
| | | 2 | 4.9643 | 3,805,560 | 536,306 | 7.0960 |
| | | 3 | 4.9639 | 3,872,147 | 546,752 | 7.0821 |
| | | 4 | 4.9681 | 3,756,711 | 531,097 | 7.0735 |
| | | 5 | 4.9648 | 3,600,182 | 507,927 | 7.0880 |
| | | 6 | 4.9694 | 3,734,811 | 528,492 | 7.0671 |
| | | 7 | 4.9661 | 3,675,843 | 515,083 | 7.1364 |
| | | 8 | 4.9672 | 3,771,143 | 531,287 | 7.0981 |
| | | 9 | 4.9718 | 3,708,926 | 523,682 | 7.0824 |
| | | 10 | 4.9666 | 3,788,234 | 534,952 | 7.0814 |
| | | 11 | 4.9695 | 3,814,125 | 538,040 | 7.0390 |
| | | 12 | 4.9673 | 3,758,823 | 530,097 | 7.0908 |
| Sept. 12 | 250 | 1 | Given above | 4,123,664 | 580,767 | 7.1004 |
| | | 2 | " " | 4,057,737 | 571,263 | 7.1031 |
| | | 3 | " " | 4,309,732 | 608,763 | 7.0795 |
| | | 4 | " " | 3,975,696 | 560,343 | 7.0951 |
| | | 5 | " " | 4,295,982 | 605,418 | 7.0959 |
| | | 6 | " " | 4,210,863 | 597,017 | 7.0531 |
| | | 7 | " " | 4,230,811 | 598,442 | 7.0697 |
| | | 8 | " " | 3,884,063 | 550,786 | 7.0518 |
| | | 9 | " " | 3,884,013 | 549,599 | 7.0670 |
| | | 10 | " " | 3,874,433 | 548,264 | 7.0667 |
| | | 11 | " " | 4,331,802 | 612,979 | 7.0668 |
| | | 12 | " " | 4,074,085 | 574,198 | 7.0952 |
| | | | | | $\bar{X}$ = | 7.0858 |
| | | | | | σ,% = | 0.29 |
| | | | | | $\sigma_{\bar{X}}$,% = | 0.059 |

* 30 second irradiation, 10 second delay, 30 second count

TABLE II. Count Data as a Function of $O^{18}$ Content

| Computed X | Measured Y | |
|---|---|---|
| $O^{18}$, %w of Total Oxygen[a] | Channels Ratio, Ch 2/Ch 4 | Y, Predicted from Regression Line[b] |
| .27970 | 7.0929 | 7.1050 |
| .29405 | 7.1687 | 7.1692 |
| .31206 | 7.2579 | 7.2497 |
| .31408 | 7.2592 | 7.2587 |
| .34994 | 7.4089 | 7.4190 |
| .35171 | 7.4429 | 7.4270 |
| .39208 | 7.6206 | 7.6074 |
| .39022 | 7.5930 | 7.5991 |
| .43527 | 7.7980 | 7.8005 |
| .45582 | 7.8860 | 7.8924 |

Pearson Product - Moment Correlation Coefficient

$r_{XY} = .99935$

$\sigma_{prediction} = \pm 0.129\%$ relative

$\sigma_{mean\ prediction} = \pm 0.041\%$ relative

[a]All but first sample prepared by addition of enriched water. All but first sample measured once. First sample, La Jolla tap water, was measured 12 times.

[b]Regression Line; obtained from observables:

$$Y = 4.4704767(X) + 5.854646$$

The last number is the ratio of backgrounds in channel 2 to channel 4 in the absence of $O^{18}$.

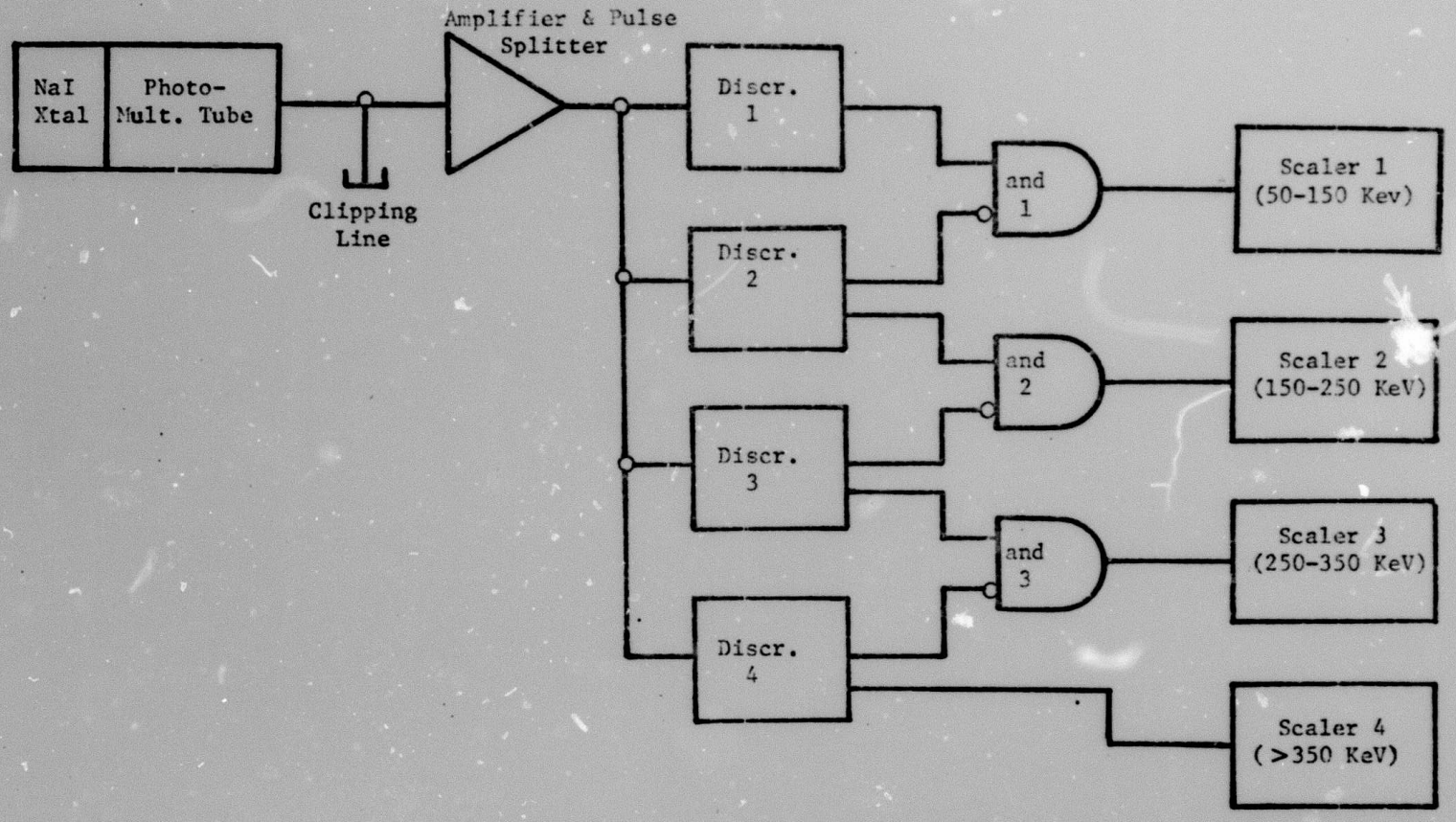


Figure 1

BLOCK DIAGRAM OF SYSTEM

Limit $\leq$ 300 Mc

## REFERENCES

1. Vials were purchased from Olympic Plastics Company, 5800 W. Jefferson Blvd., Los Angeles, 90016.

2. H. R. Lukens, D. M. Fleishman, and R. L. Bramblett, "Determination of U-235 in Highly Enriched Fuel Sticks", Proceedings of Conference Internationale sur les Tendances Modernes de L'Analyse par Activation, Centre European Nationale, Sodet, Oct. 1972, to be published in J. Radio Analytical Chemistry.

3. The method used to evolve $CO_2$ gas for measurement in the mass spectrograph was to heat about 50 mg. $H_2O$ together with several hundred mg of $HgCl_2 + Hg(CN)_2$ as discussed by D. Rittenberg and L. Pontecorvo, "A Method for Determination of the $O^{18}$ Concentration of the Oxygen of Organic Compounds", International Journal of Applied Radiation and Isotopes 1, 208-214 (1956). The reagents are sealed together with the water drop in a highly evacuated quartz bomb at liquid air temperature, then heated at 500°C for half an hour, to release $CO_2$. HCL is removed by a quinoline trap.