

A Pilot Experiment on Acid-Alkali-Acid (AAA) Pretreatment for Accelerator Mass Spectrometry (AMS) Measurement of ^{14}C at KOMAC

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1. Introduction

Accelerator mass spectrometry (AMS) is an ultra-sensitive technique for measuring low concentrations of natural isotopic abundances of both radioisotopes and stable isotopes. The sensitivity of AMS measurement ranges between 10^{-12} and 10^{-16} . The main advantage of AMS compared to traditional mass spectrometry methods is the use of smaller samples (mg and even sub-mg size). AMS can be applied in the technique of ^{14}C dating in the fields of archeology and geology. The archetypal example is ^{14}C which has a half-life of 5730 years and an abundance in living organisms of 10^{-12} relative to stable ^{12}C . Using AMS, the radiocarbon age of a sample less than 10,000 years-old can be determined with a precision of 0.5% in a few minutes using a mg or less of sample [1,2].

Here, we conducted a careful investigation of chemical pretreatment to determine the best method of collecting radiocarbon reflecting the sample age.

2. Methods and Results

2.1. Materials and instrumentation

HCl and NaOH were purchased from Sigma-Aldrich (St. Louis, USA). All AMS pretreatment instruments for sample combustion, CO_2 collection and graphitization were moved from Seoul University. Photograph of all instruments was shown in Fig. 1.

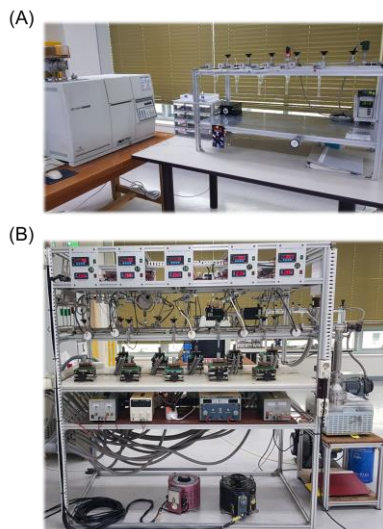


Fig. 1. Photograph of (A) the EA, the CO_2 trap and (B) the reduction system, employed for combustion, CO_2 collection and graphitization of organic samples for KOMAC.

2.2. Acid-Alkali-Acid (AAA) treatment

De Vries et al. [3] in 1954 reported sample pretreatment, using hydrochloric acid (HCl), sodium hydroxide (NaOH) with water washes between the three steps to remove contamination by carbonates and humic acids. Such a pretreatment is often called an AAA treatment and is a popular method [4]. We described the detail AAA procedure used at KOMAC as follow:

1. Stirred in 1 M HCl solution at 80 °C for 2 hr
2. Washed with water three times to neutral pH
3. Stirred in 1 M NaOH solution at 80 °C for 1 hr
4. Washed with water three times to neutral pH
5. Stirred in 0.5 M HCl solution at 80 °C for 1 hr
6. Washed with water three times to neutral pH
7. Stirred in water at 80 °C for 0.5 hr
8. Dried in a 60 °C oven overnight

2.3. Combustion and CO_2 collection

The AAA treated samples are combusted to release CO_2 . Commercially available elemental analyzer (EA) instrument is used to isolate the CO_2 and N_2 . The CO_2 was at first trapped from the waste line cryogenically by liquid nitrogen, and transferred to the reduction system [5,6].

2.4. Graphitization

After the separation, the CO_2 gas is reduced, in the presence of iron powder as catalyst, to carbon in the form of graphite. The reduction is most commonly done under hydrogen [5,7].

2.5. AMS analysis

The $^{14}\text{C}/^{12}\text{C}$ ratio of each sample was measured by the AMS using the 6-MV tandem accelerator at the Korea Institute of Science and Technology (KIST). Result is presented in Table I.

Table I. The $^{14}\text{C}/^{12}\text{C}$ ratio of anthracite and oxalic acid II samples measured by 6-MV tandem accelerator at KIST.

Sample	$^{14}\text{C}/^{12}\text{C}$ ratio
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Anthracite-1	2.61.E-15
Anthracite-2	2.15.E-15
Anthracite-3	5.52.E-14
Anthracite-4	2.24.E-15
Oxalic acid II-1	1.57.E-12
Oxalic acid II-2	1.55.E-12
Oxalic acid II-3	1.53.E-12
Oxalic acid II-4	1.53.E-12

The $^{14}\text{C}/^{12}\text{C}$ ratio of anthracite is around 10^{-15} . This value is within the expected range, since anthracite is the commonly used material for background determination [8]. However we found the unexpected value from Anthracite-3 sample ($\sim 10^{-14}$). So we are trying to trace the pretreatment procedure of Anthracite-3 sample to retain a constant background level during ^{14}C analysis. In case of Oxalic acid II, standard material, showed an expected ratio of around 10^{-12} [9].

3. Conclusions

We prepared 8 samples (background material, $n = 4$; standard material, $n = 4$) and measured the $^{14}\text{C}/^{12}\text{C}$ ratio using AMS system at KIST. However, we still have not reached satisfactory results. In order to get reproducible results of AMS measurement, we are continually investigating the most stable pretreatment method and are optimizing the procedure.

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