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Mass spectrometric detection of radiocarbon for dating applications

H.-A. Synal *, T. Schulze-König, M. Seiler, M. Suter, L. Wacker

ETH Zurich, Laboratory of Ion Beam Physics, Building HPK, 8093 Zurich, Switzerland

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ABSTRACT

Radiocarbon is still the most important nuclide measured by accelerator mass spectrometry (AMS). The related capabilities for dating and tracer studies are eminent not only in archaeology but also drive important applications in the earth and environmental sciences as well as in biomedical research. So far, standard mass spectrometric systems have not been capable of radiocarbon dating because of interfering molecular isobars which, however, can be completely eliminated in charge changing processes at high ion beam energies (MeV) [1,2]. Here, we present a novel type mass spectrometry system for radiocarbon analyses. Radiocarbon dating was performed using 45 keV ^{14}C ions from the ion source and a molecule dissociation unit kept at ground potential. This proof-of-principle experiment demonstrates for the first time the feasibility of mass spectrometric radiocarbon dating without an accelerator. The results obtained will be the basis of an optimized design for a radiocarbon dating instrument comparable in size, complexity and cost to standard mass spectrometers.

1. Introduction

The long half-life of radiocarbon (5700 yrs) makes direct detection of ^{14}C atoms/ions more sensitive by orders of magnitude than decay counting. However, to reach the sensitivity necessary for radiocarbon dating, mass spectrometric techniques require an abundance sensitivity of $1:10^{15}$ for the rare nuclide in the presence of the most abundant isotope, ^{12}C. In general, atomic as well as molecular isobaric interferences limit the capability of mass spectrometers. By using negatively charged ions, the atomic isobar ^{14}N can be eliminated [1–3] and the remaining challenge is to efficiently remove the molecules of mass 14 ($^{12}CH_2$, ^{13}CH and $^{7}Li_2$). Accelerator mass spectrometry has overcome this obstacle by stripping electrons off such molecules at MeV energies to reach charge states 3+ and higher, where no bound states exist [4]. It has, however, been shown that molecules in charge state 1+ can be destroyed in multiple ion collisions with Ar or N_2 gas [5,6]. For these gases, the maximal 1+ charge state yield for ^{14}C (≈55%) is reached at stripping energies of 450 keV and thus much smaller accelerators could be used reducing the overall size of the AMS spectrometer [5,7]. Today, the most compact AMS systems utilize 1+ ions and operate at 200 kV terminal voltage using conventional high voltage power supplies [8].

A further reduction of the ion energy would allow to build instrumentation for radiocarbon detection comparable in complexity to standard isotope ratio mass spectrometers. The challenge is still to eliminate the interfering molecules. The cross-section for molecule dissociation in ion–gas collisions is the most critical parameter. But the process of molecule destruction is also accompanied by a distortion of ion beam characteristics. Ion–gas collisions cause energy loss and angular straggling, which makes it difficult to transport the ions with high transmissions through a mass spectrometer. These obstacles become severe for low ion energies, when medium–heavy gases such as Ar or N_2 are used for stripping.

Recently, we have measured dissociation cross-sections for mass 14 molecules in N_2 and helium in the energy range of 80–250 keV [9]. Whereas in N_2 cross sections go down at ion energies of ≈100 keV, the He data remain fairly constant. This has encouraged us to further explore the properties of He at energies, which can be directly extracted from an ion source. In such a case, the acceleration stage necessary in any AMS spectrometer can be omitted.

Our primary objective was to construct a He stripper gas canal to reduce the molecular beam component sufficiently enough to reach the sensitivity required for radiocarbon dating. The stripping yield for 1+ carbon ions in He has been reported to be about 60% at energies of about 100 keV [10,11]. Due to reduced angular straggling of carbon ions in the light He stripping gas, a high transmission was thought to be achievable even at moderate angular acceptance angles of the ion optical components. Based on this, we set up a proof-of-principle experiment to demonstrate the feasibility of radiocarbon dating using a mass spectrometric system without an accelerator.

* Corresponding author. Tel.: +41 44 633 2027; fax: +41 44 633 1067.
E-mail address: synal@phys.ethz.ch (H.-A. Synal).

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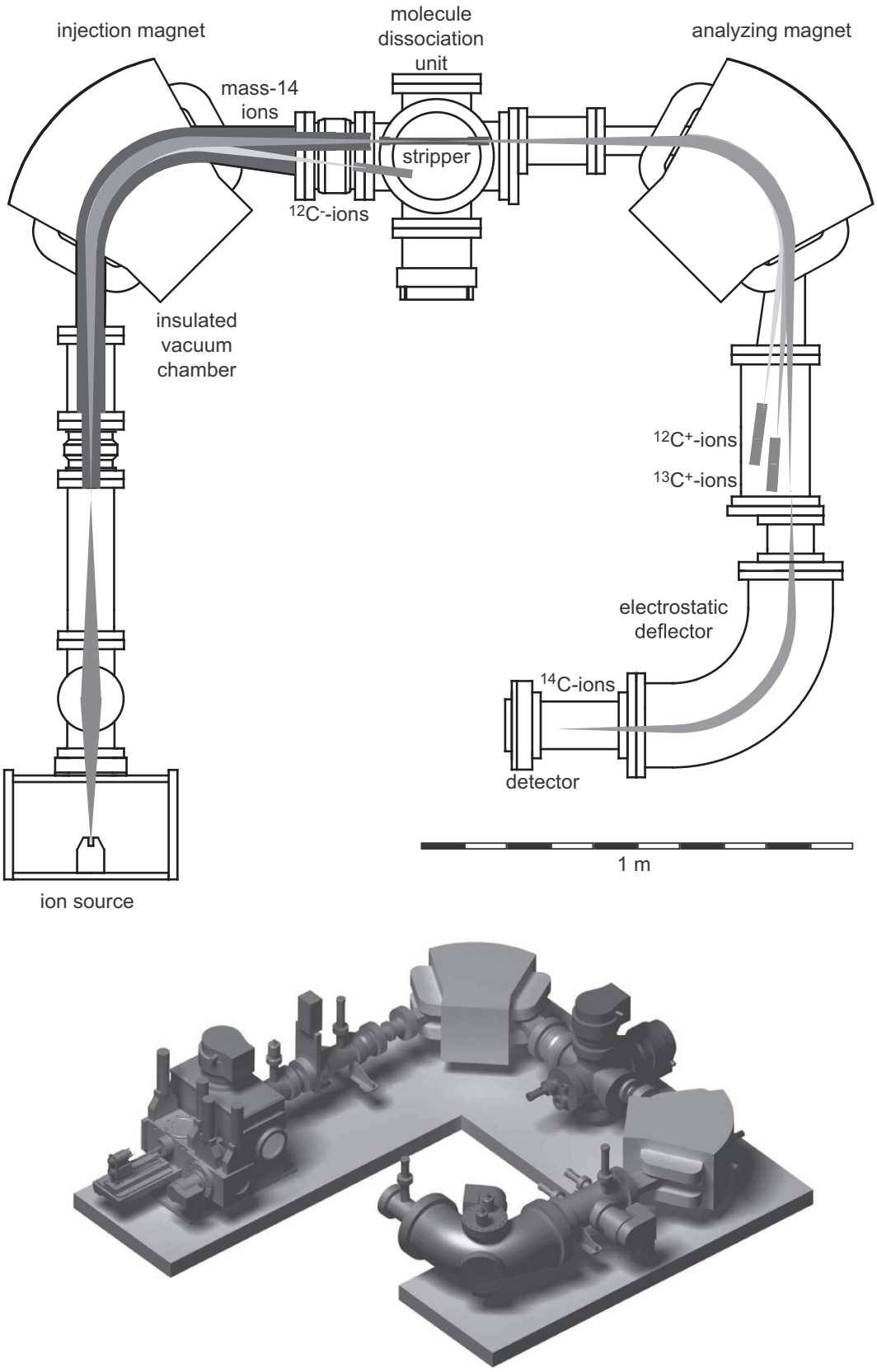

Fig. 1. Experimental setup as it was used for the first mass spectrometric ^{14}C dating measurements. The overall dimensions of the entire system are 2×2.5 m^2. The molecule dissociation unit is located in between the two magnets. The electrostatic deflector is the last filter element in front of the detection system. The section in which the energy of the ions can be changed by the beam switching system is marked in dark grey. The design view at the bottom of the figure is shown not to scale.