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AMS measurement technique after 30 years: Possibilities and limitations of low energy systems

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ABSTRACT

A brief summary of the development of AMS over the last 30 years is given and major development steps for the measurement technique are described. With the appearance of compact low energy AMS systems 10 years ago, a new category of AMS instruments has been introduced. This has resulted in a boom of new AMS facilities with more than 20 installations over the last 5 years. But low energy AMS is not limited to radiocarbon only and there is a great potential for ^{10}Be, ^{26}Al, ^{129}I and actinides measurements at compact AMS systems. The latest developments towards the low energy limit of AMS resulted in two new types of systems, the NEC Single Stage AMS and ETH MICADAS operating with terminal voltages of about 200 kV only. These systems have enormous impact, not only on the use of AMS in biomedical research but also on radiocarbon dating. Precision limits of radiocarbon dating will be discussed for the MICADAS type instruments.

1. Evolution of the AMS technique

From its advent more than 30 years ago, accelerator mass spectrometry (AMS) has made tremendous progress. This is true not only for the large variety of possible applications exploiting the unique analytical capabilities of AMS, but also for the measurement technique itself. However, its fundamental principles developed in 1977 in connection with tandem accelerators [1,2] are still the basis of state-of-the-art AMS instrumentation. In contrast, the initial impact of cyclotron based AMS systems [3] on the direct detection of long-lived radionuclides have lost importance except for noble gases Ar and Kr, which cannot been measured with tandem accelerators [4,5]. Only one experimental cyclotron based AMS system [6] exists, which to date has not reached the state of routine operation. Shortly after the discovery that radiocarbon measurements are possible with tandem accelerators, it was realized that the instrumentation initially used would not be able to cope with the requirements of this new powerful technique. Neither the necessary sample throughput nor the desired overall precision could be reached with the instruments available at that time. Already at the first AMS conference in 1978, Kenneth Purser presented the first proposal for a dedicated AMS system [7]. The first two spectrometers of this type were produced by General Ionex Corporation and commissioned at the Universities of Arizona and Toronto [8]. Later, such systems were acquired by Oxford University [9], Nagoya University [10] and Gif-sure-Yvette [11]. Beside the rise of commercial instruments, several nuclear physics accelerator laboratories have added dedicated AMS instrumentation to their beam lines. Whereas laboratories with large accelerators (HVEC FN and MP Tandems, NEC Pelletrons) only modified their equipment (Rochester [12], McMaster University [13], Philadelphia [14], Chalk River [15], Munich [16], Canberra [17], Rehovot [18]), laboratories hosting EN tandems made more fundamental changes to their installations: Zurich [19], Utrecht [20], Uppsala [21], Aarhus [22], Erlangen [23] and Lower Hutt [24]. In particular, Zurich [19] followed the ideas as outlined in the first AMS proceedings and established a benchmark system for high precision ^{14}C measurements during the early times of AMS [25].

Triggered by the need of highly precise radiocarbon dates in connection with the World Ocean Circulation Experiment WOCE [26] an AMS system based on a 3 MV Tandetron accelerator was proposed by Kenneth Purser [27,28] and installed in 1991 at the Woods Hole Oceanographic Institution [29]. After 3 years of operation, the system had reached routine precision of better 5‰ [30]. Similar systems followed in Groningen [31], Kiel [32], Nagoya [33], JEARI [34], Seoul [35], Jena [36] and Oxford [37]. Intensified routine operation of these systems resulted in improved performance. The Kiel facility, for example, became able to deliver uncertainties of radiocarbon results at the 3‰-level under routine operation conditions. Thus, this type of instrumentation was the most favored in the 1990s and High Voltage Engineering Europa (HVEE) who had acquired the patents from Kenneth Purser was dominating the market of commercially built AMS instruments at that time. In contrast to all other AMS systems, which are using sequential injection sys-

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tems either with slow or fast beam switching sequences, the HVEE systems were equipped with a re-combining magnetic inflection spectrometer [28] providing achromatic injection conditions [38]. Parallel injection had originally been proposed by Lobb et al. [39] for the AMS system at the MacMaster University. Over many years, a controversial discussion on the best-suited AMS injection system went on [40] but both types of systems survived. However, the majority of the AMS systems worldwide have sequential systems, not at least due to their capability to inject a wide variety of ion beams. Today, also the HVEE 3 MV Tandetron systems can be furnished with sequential injection lines. The first system of this type was installed in Lecce, Italy [41] followed by the new AMS system in Florence, Italy [42]. Both systems will be used in connection with ion beam analysis methods.

In July 1994, the original patent on the fundamental process to eliminate molecular interferences [43] expired and National Electrostatics Corporation (NEC) entered the market of commercially built AMS systems. At first, NEC focused on systems based on their 5 MV Pelletron accelerator and their first installations were made for the University of Tokyo [44], the National Institute for Environmental Studies, Tsukuba, Japan [45] and the Tono Geoscience Center, Gifu Japan [46]. In parallel, the installation of the first 3 MV Pelletron based dedicated AMS system in Vienna [47] was accomplished in 1996 and more recently, systems based on 3 MV Pelletron accelerators were installed at the University of Arizona [48], Artemis Saclay [49], CIRCE AMS in Caserta, Italy [50] and the Institute of Accelerator Analyses (IAA), Fukushima, Japan [51].

2. Dedicated facilities based on large accelerators

It had always been the question of which type of accelerator would be optimal for dedicated AMS systems. Of course, there is no easy answer and the best-suited accelerator depends on the purpose of the facility. A pure radiocarbon dating system has different requirements than a multi-purpose AMS system covering a range of AMS nuclides for which substantial isobar suppression capabilities are needed. FN accelerators reaching terminal voltages of 10 MV seemed to be well suited as basis of versatile multi-purpose AMS systems. At Lawrence Livermore National Laboratory, the CAMS–AMS facility [52,53] was set up using the FN tandem purchased from Washington University. At Purdue the existing FN facility was converted into an AMS system [54], and at the Australian Nuclear Science and Technology Organisation the ANTARES–AMS facility was built around the former FN tandem of the Rutgers University. Nevertheless, the early AMS systems in Philadelphia [14], Seattle [55] and the cradle AMS instruments at McMaster University and at Rochester University were closed down. Large Pelletron and MP tandem accelerators with terminal voltages ranging from 14 to 25 MV (Canberra, Munich, Oakridge [56]) are still used for AMS measurements, predominantly for heavier nuclides which suffer from abundant nuclear isobar interferences. In general, the effort to operate and maintain such large accelerator facilities is too high to justify it with an AMS program only and beam time at these facilities is shared with other activities. Experimental AMS work at large facilities has been conducted in Brazil [57,58] and Argentina [59,60].

3. The golden rule of AMS and the advent of compact AMS instruments

To detect long-lived radionuclides at natural concentrations mass interferences of any kind have to be filtered out. Apart from isobaric nuclides, molecular ambiguities have to be removed. Over the first 20 years of AMS, it had been the golden rule to use charge states 3+ or higher to eliminate molecular ions, because molecules do not have bound states with such electron configurations. Although experiments in the early 1980s had already shown [61,62] that beam intensities of molecular ions in charge states lower than 3+ decrease exponentially with increasing density of the stripper medium, it was not believed that detection of radiocarbon at natural levels could be performed using charge states 1+ or 2+. Systematic investigations of the processes involved revealed that cross sections for dissociation of mass 14 molecules ($^{12}CH_2$, ^{13}CH) are sufficient to reduce their interference with radiocarbon over 11 orders of magnitude at quite moderate stripper densities [63]. This has made it possible to use charge state 1+ and to reach detection levels as are required for real dating applications. In 1998, the first system using charge state 1+ became operational [64] and it was demonstrated that this technology is capable to compete with traditional AMS systems using charge states 3+ or 4+ [65]. The new way to eliminate molecular interferences has the great advantage that the maximal yield for charge exchange to 1+ can be reached at energies of about 500 keV, which is much lower than the energy for 3+ or 4+ maximal yield at 2.5 MV or 6.5 MV terminal voltages, respectively. Consequently, AMS systems based on this principle are more compact with smaller accelerators and can be produced and operated at much lower cost than traditional systems. The development of compact AMS systems for radiocarbon was undertaken at ETH in collaboration NEC. NEC has used these developments to commercialize AMS systems based on the new technology. Such compact AMS systems were set up first at the University of Georgia, Athens [66] followed by systems in Poznan, Polen [67], Irvine [68], Accium, Seattle [69], Paleo Labo Co, Japan [70], CHRONO Center, Belfast [71], at the IAA, Fukushima, Japan [51] and Peking University, China [72]. The obvious advantages of the compact systems have also been realized by HVEE which now has proposed its own compact multi-nuclide AMS system [73]. The first instrument of this type was installed at University of Seville in Spain in 2005 [74]. In contrast to the compact NEC systems, which are optimized for radiocarbon, the compact HVEE system has the option to analyze beams up to plutonium.

4. AMS at very low energies

Shortly after proof of principle of the new method to eliminate molecular interferences, the question of the low energy limit of AMS arose. Detailed measurements of molecule dissociation cross sections [75] revealed that even at energies as low as 200 keV, the cross sections are sufficiently high to achieve suppression of molecular interference down to levels where dating measurements can be performed. Stripping yields for the 1+ charge state are still approximately 50% [64] and thus high enough for efficient radiocarbon detection. Traditional accelerator systems are not necessary anymore to obtain ion energies in these ranges. At ETH we have designed a vacuum insulated high-voltage platform in a tandem configuration fed from a commercial high voltage power supply and hosting a charge exchange channel that provides sufficient area density of a stripper gas to eliminate molecular interferences. Based on initial tests of this device [76], a prototype dedicated radiocarbon dating system (MICADAS) [77] was built and became operational in 2004. The MICADAS type system has a footprint of $2.5 \times 3\ m^2$ only and is therefore the smallest AMS instrument capable of high performance radiocarbon measurements. In parallel, NEC has developed a single stage open stack AMS system (SSAMS) [78]. These systems operate either with the injection part of the system or the analyzing branch including the detection system at elevated voltage. The simple design, the related reasonable system price and the excellent performance of this type of instruments have resulted in quite a number of new AMS installations after its first appearance in Lund [79].