

## **An Introduction to Hypersensitive Analysis by AMS**

**C t lin Stan-Sion**

*National Institute for Physics and Nuclear Engineering, Department for Applied Physics, Bucharest*

**Mihaela En chescu**

*National Institute for Physics and Nuclear Engineering, Department for Applied Physics, Bucharest*

**Cristina Cîrtoaje**

*University "Politehnica" of Bucharest, Physics Department, Bucharest, Romania*

**Emil Petrescu**

*University "Politehnica" of Bucharest, Physics Department, Bucharest, Romania*

### **Abstract:**

*Rare radio-isotopes can give valuable information in different scientific areas, from medicine to cosmic rays studies. This is the reason why Accelerator Mass Spectrometry (AMS) has been developed to perform analyses at highest sensitivities, able to detect radio-isotopes concentrations where other methods fail. AMS with  $^{14}\text{C}$  performs precise dating of objects of biologic origin using only very small samples reducing relic destruction.  $^{10}\text{Be}$  is a long lived isotope used in historical studies of samples from the last ten million years. It can also be used in geology and its high analysing sensitivity allows measuring the  $^{10}\text{Be}$  produced directly inside the rock by the cosmic rays. From only 1 ml of blood AMS can detect the aluminium ( $^{26}\text{Al}$ ) absorption in different human organs.  $^{36}\text{Cl}$  is a good atmospheric circulation tracer that can be detected by AMS.  $^{129}\text{I}$  has a reduced specific activity but high volatility and can be used efficiently as a tracer of nuclear pollution. The paper will present the method of AMS and will give the reader an introduction into the possibilities of using this analysing method in different research activities.*

**Keywords:** Accelerator mass spectrometry, radio-isotope.

### **1. Introduction**

The modern society, through its permanent development, encourages rapid absorption of the newest scientific progress, encouraging discoveries with worldwide applications in different human fields. As such, scientific progress in regard to atomic and nuclear physics is of major importance. It contributes to the permanent development of the newest methods for the investigation of our "universe". Among these methods, the analyzing techniques are essential in modern microscopic investigation of matter. Accelerator Mass Spectrometry (AMS) is a very unique and special method of selecting and individually counting atoms of a certain kind. It has the highest analysis sensitivity known today:  $10^{-16}$  (ratio: isotope/element). This sensitivity is equivalent with the possibility to select and count one single type of atom from one million of billions of other types of atoms. The applications of AMS are meant for many different fields: medicine, biology, environmental physics, geology, forensic science, nuclear pollution, solid state physics, etc.

## 2. AMS General Description

AMS is measuring the rare isotopes concentration in a sample material. As mentioned above, it has a very high sensitivity, being able to separate and register one single atom from more than one billion of millions of other atoms. A typical AMS facility, as presented in Figure no. 1, consists of four major parts: the ion injector (containing the ion source); the tandem accelerator; the high energy analysing systems and the particle discrimination and detection system. As it will be described in more detail below, these parts contain a series of mass filters that produce the desired separation and at the end they are able to produce the high sensitive analysis of certain atoms.

Basically, this kind of analysis is performed by producing an ion beam from the sample material itself. This beam is produced by the ion source and afterwards it is accelerated to high energies enough for using efficiently the electromagnetic devices that separate the desired ions from the rest of the ions produced from the sample material. Finally, a particle detector is counting the selected ions one by one and as a result, the concentration of the enquired element in the analyzed sample can be determined.

**Figure no. 1. Schematic diagram of an AMS- facility**

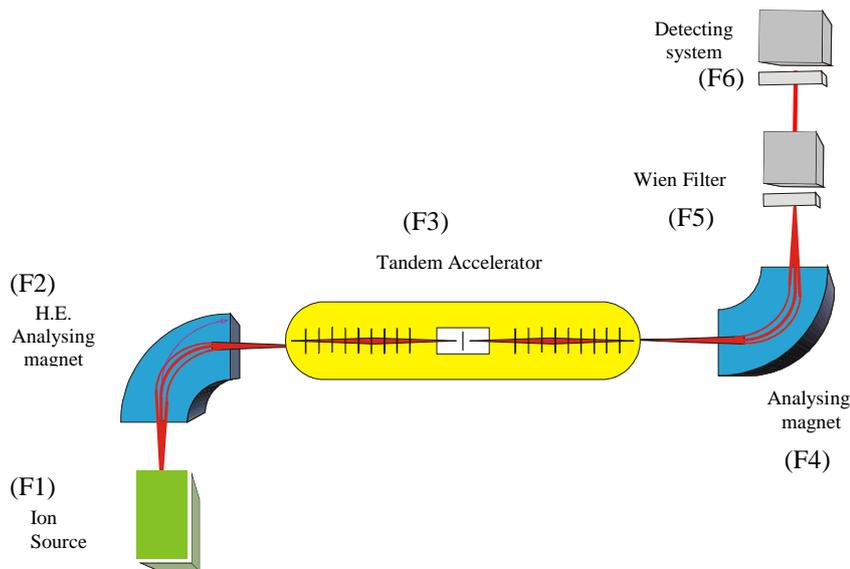
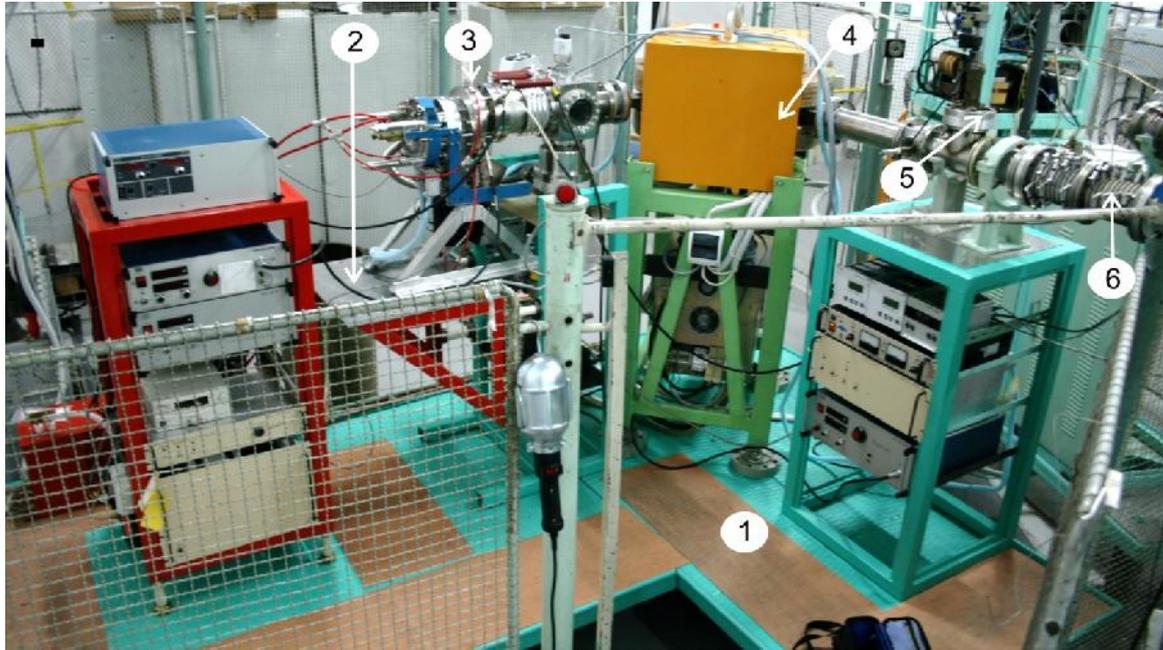


Photo no. 1 is showing the injector deck, where the ion source is located. Fig.2 shows the schematic drawing of the sputter ion source and its functioning. It represents the first AMS filter (F1), since it produces as output only negative ions (see fig.1). Positive molecular ions or neutrals are also produced but will be removed from the beginning and in this way, the perturbing interferences are avoided. In order to get an adequate form for their use in ions sputtering source, the probes to be analyzed are first chemically prepared. The AMS sample should have a solid substance, with good thermal and electrical conductivity. Inside the ion source, an accelerated beam of positive ions of  $^{133}\text{Cs}$  is focused and bombards the sample to be analyzed. By a sputtering process, all kinds of atoms will be scattered out from the sample material. However, only the negative ions will be extracted from the ion source. The extraction energy is about 25 keV. These negative ions can be further accelerated by the tandem accelerator on its first acceleration path up to the positive loaded HV terminal.

The second filter is the  $90^\circ$  double focusing analysing magnet (F2), located on the injector deck, too (see Photo no. 1). By passing through this magnet, the negative ions ( $q = -1$ ) extracted from the ion source will be selected according to their mass value with a resolution of  $\Delta m/m = 1/250$ . Then, before entering into the tandem accelerator, the selected ions species will be further accelerated up to about 70 keV. The acceleration tubes (NEC type) are located at the injector exit.



**Photo no. 1. The new upgraded ion injector deck of the AMS facility in Bucharest:**

1) Injector deck polarized -100 kV, 2) second platform, polarized -30 kV with respect to the injector deck, 3) the 40 NC-SNICS, 4) the analyzing magnet, 5) Slits and retractable Faraday Cup, 6) pre-acceleration NEC tubes.

This energy increase is necessary to reduce the source emittance and in this way to match the entrance acceptance of the tandem accelerator. The emittance is a measure of disorder of the accelerated particles inside the ion beam and is given by the ratio: transversal momentum/ axial momentum ( $p_t/p_x$ ).

When the 70 keV negative ions ( $q = -e$ ) enter the tandem accelerator, they are further accelerated by the potential difference between the input of the system and the terminal of high voltage (typically, from  $V_p = 0$  to  $V_t = 8$  M V). At the central terminal position, the accelerated ions have to pass through the stripper foil (carbon foil, 6  $\mu\text{m}/\text{mg}$ , or gaseous medium) and loose part of their exterior electrons by the stripping process in the foil. In this way, the negative ions will change to positive charge, with different values ( $q = 2+, 3+, 4+, 5+, 6+, 7+, \dots$ ). The charged values depend on the incident energy on the foil. Molecules passing through the terminal stripping foil are exploded and therefore are removed from the ion beam. That is the reason why the stripper foil is considered to be the third filter (F3). After the stripper foil, the now positively charged ions will be repelled by the positive high voltage terminal. They will be accelerated in the second half of the accelerator system, down to the ground potential value and up to the exit of the tandem accelerator. At this location, the energy will be about 48.07 MeV ( $E_{\text{Tandem}} = U(q+1) = 8 \times (5+1) = 48$  plus 70 keV).

At the tandem exit, after ion focusing and a few meters of free flight, a large and powerful magnetic analyser is performing the high resolution of mass separation. This is the fourth filter (F4).

Since the magnetic field cannot separate particles with the same p/q ratio a Wien velocity filter can be used to separate the ions according to their velocity and in this way remove the magnetic degeneracy. The velocity filter is located in between the HE analysing magnet and the detection system. This is the fifth filter (F5).

Finally, the AMS facility ends with the particle detection system, which is the last filter (F6).

However, there is a major difficulty in the AMS analysis: because of the very small concentrations to be determined, the intensity of the corresponding produced ion beam intensity is only of a few ions per minute. There is no way of measuring this extremely low beam current intensity, excepting in the final detector system. Moreover, the transporting of such a low intensity ion beam through a long (more than 35 m) and complex beam transport system that preserves high vacuum is not directly possible.

First, one has to use an ion beam with a measurable electric intensity to tune the beam transport through the entire AMS facility. This pilot beam will be produced from the substrate of the sample material. For example,  $^{26}\text{Al}$  will be measured in a substrate sample of  $^{27}\text{Al}$  and  $^{27}\text{Al}$  will be the pilot beam,  $^{129}\text{I}$  will be measured in a substrate sample of  $^{127}\text{I}$ , with this ion used as pilot, etc. The pilot beam will be used to adjust the focusing of ion optics for the entire ion beam transport up to the entrance into the detection system.

Once the tuning of the AMS analyzing system was optimized, the rare isotope beam can be extracted from the ion source (the analysing magnet on the AMS injector will select blindly the microscopic beam). If the condition of constant rigidity is preserved, this beam will be transported exactly on the same path with that of the pilot beam, all the way down to the detection system. This condition is given by the relation:

$$(\text{BR})^2 = 2 \text{EM}/\text{q}^2 = \text{constant} \quad (1)$$

Where M, q, E represent the mass, the charge state and the energy of an ion, B and R, the magnetic intensity and the curvature radius of the magnet bending respectively. Practically, one has to maintain most of the tuned values for the devices from the AMS facility unchanged, with two exceptions. These are: 1) the value of the magnetic analyser on the injector deck and 2) the value of the tandem terminal voltage.

Let us consider the  $^{26}\text{Al}/^{27}\text{Al}$  concentration measurement as an example. The pilot beam will be of  $^{27}\text{Al}$  ions. According to the constraint given by the relation (1), the analyzing magnet of the ion injector, together with the terminal voltage of the tandem accelerator has to be changed to new values:  $B_{26} = B_{27} \times (26/27)^{1/2}$ . Furthermore, since the energy gained in the accelerator is related to the terminal potential value by the relation  $E = U(q+1)$ , the change for the terminal voltage will be  $U_{26} = U_{27} \times 27/26$ , if  $q_1 = q_2$ . After this change, the detector will be registering the pure  $^{26}\text{Al}$  ions and the interference from  $^{26}\text{Mg}$ . However, this interference can be removed by the velocity Wien filter.

AMS is usually a relative analysing method and requires standard samples. In this way one can obtain absolute values of the measured concentrations.

### 3. Several Applications of AMS

The AMS method is used in many and various applications [1, 2, 3] and because of its exceptional analysis sensitivity it managed to open new applications. One of the most important aspects of Accelerated Ions Mass Spectrometry is its potential to give a good understanding of natural phenomena and transformations suffered by the Earth along its long history. In this respect, radio-nuclides with long life represent a valuable instrument. They represent very good tracers or markers of processes and are not affected by interferences from the environment. Since long life radio-nuclides have also very low concentrations, these cannot be measured directly by disintegration. Hence the advantage of AMS that measures the radio-nuclides by counting the atoms individually.

As an example, for the efficiency of the AMS, we shall consider the measurement of the  $^{14}\text{C}$  quantity contained in a hair wire with the length of 2 cm. Such a hair wire contains usually about 1mg of carbon in the form of the protein compound called keratin. The carbon milligram contains  $5 \times 10^{19}$  stable  $^{12}\text{C}$  atoms. From the natural concentration ration  $^{14}\text{C}/^{12}\text{C} = 1.2 \times 10^{-12}$ , it results that the hair contains  $6 \times 10^6$  radioactive atoms of  $^{14}\text{C}$ . The half time of the radio-nuclide is 5730 years. It needs about an hour for a single atom to disintegrate and emit the specific radiation that can be detected with a specialized apparatus. When using the AMS method, with an efficiency of 10% for  $^{14}\text{C}$  detection, the result is that in the same interval of one hour  $6 \times 10^5$  atoms will be counted and the registering statistics is greater by about six orders of magnitude in comparison with the measurement of radioactive decay. This difference is even more relevant for the radio-isotopes with longer half times.

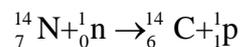
Today, after about 25 years of scientific and technical development, from the 100 radio-nuclides that can be used for AMS, the following ones are currently used:  $^2\text{H}$ ,  $^3\text{H}$ ,  $^{10}\text{Be}$ ,  $^{14}\text{C}$ ,  $^{16}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{41}\text{Ca}$ ,  $^{44}\text{Ti}$ ,  $^{53}\text{Mn}$ ,  $^{59}\text{Ni}$ ,  $^{60}\text{Fe}$ ,  $^{63}\text{Ni}$ ,  $^{129}\text{I}$ ,  $^{236}\text{U}$ ,  $^{244}\text{Pu}$ , etc.

In the last decade, there is a large list of applications of the AMS in geology (subterranean water tracers and erosion rate determinations of earth crust), in medicine (in vivo tracers of human bio-kinematics and cell level changes), in astrophysics (material composition of extraterrestrial and meteoric studies, the studies of solar activities), nuclear physics (measurements of reaction sections and life times of radio-nuclides produced by cosmic and solar radiations), paleontology (studies of human fossils), pale-climatology (climate evolution determinations), pale-geomorphology (relief evolution in the past of our planet and geo-morphological processes), atmosphere physics (the stratosphere-troposphere changes and the study of the dynamics of the atmosphere), contamination tracing (produced by nuclear power stations) around the country, current production (in areas like metallurgy, construction materials, fine mechanics, semiconductors, composites, pure and ultra-pure materials).

In the next chapters, we will present some of the applications listed above in more detail. However, it has to be emphasised that each application has been developed nowadays to such an extent that only a short introduction to those subjects serves sufficiently the purpose of this paper. For a more detailed description of the individual studies, the interested reader is advised to follow the indicated bibliography.

### 3.1. $^{14}\text{C}$ Radio-isotope

$^{14}\text{C}$  has a half time of 5370 years. Its usage with the AMS method allows exact dating of objects for a very long period in our history. It is produced in the earth's atmosphere by the reaction of high energetic neutrons with nitrogen:



The isotope forms easily the carbon dioxide which is then assimilated by plants, similarly with the stable isotope  $^{12}\text{C}$ . Then,  $^{14}\text{C}$  will be spread over air, plants, food, etc. and it will be found in the tissue composition and in different organs. At each time in our history, the ratio  $^{14}\text{C}/^{12}\text{C}$  has a certain value. If an organism dies, the assimilation of carbon dioxide stops and the ratio between the isotopes  $^{14}\text{C}$  and  $^{12}\text{C}$  starts decreasing. This is valid for plants, humans or living animals and one can measure the time period from the moment when the carbon assimilation was stopped. AMS measurements performed on samples from the wooden substance of secular tree rings determined the values of the  $^{14}\text{C}/^{12}\text{C}$  ratio along the history of the past 9000 years. This time curve is correlated with the existence of major processes on Earth and with periodic solar activities. The  $^{14}\text{C}$  curve is currently used for the interpretation of carbon dating.

A recent scientific study of the presumed shroud of Christ from Turin [1] pointed out the advantages of the AMS method as the only method capable of using an insignificant mass of sample material for the most sensitive and precise analysis. After considerable negotiations with the church authorities, three AMS laboratories were chosen (from Arizona, Oxford and Zürich) to make the measurements with radiocarbon on a small piece (50 mg) from the presumed shroud of Christ from Turin. The three laboratories found a good correspondence between the experimental results for the age of the shroud and other three probes of known ages. It was concluded that the shroud came from the Middle Ages (1260-1390 AD), the result having a 95% confidence limit. This result is also in accordance with the opinion of art history experts stating that the shroud is a medieval icon rather than one from the time of Christ.

Similar AMS studies identified that the iced man from the Tyrol Alps is from the Late Neolithic (3120 - 3350 BC) rather than from the Bronze Age [4].

Recent  $^{14}\text{C}$  AMS dating produced a real revolution in pale-anthropology. Measurements were possible on collagen and even amino acids. Many of the important dates in human history were corrected.

Also, AMS measurements are currently made by the World Ocean Community. This organization launched a global program to determine  $^{14}\text{C}$  distribution in planetary oceans to understand the major circulation of deep and surface sea currents. From the content study of the  $^{14}\text{C}$  in foraminifers, microscopic molluscs and various species living in oceans on different depths, one could determine circulating diagrams of ocean water in the past. In a sediment probe, the ratio  $^{14}\text{C}/\text{C}$  from plankton foraminifers gives a measure of the difference of the ratio between the surface water and the deep water at the sedimentation moment. This difference can be interpreted as an age difference between the surface and deep waters. By using the AMS method, the conclusion of these results was that the deep water of the Atlantic Ocean was twice as old in the Ice Age as it is in

our day. This involves a slower surface circulation of the deep waters in the Ice Age than now [5].

### 3.2. $^{10}\text{Be}$ Radio-isotope

$^{10}\text{Be}$  ( $T_{1/2}=1.6 \times 10^6$  years) is produced in the atmosphere by spallation reactions induced by secondary protons and neutrons (the result of galactic cosmic radiation) on nitrogen or oxygen. It has one stable isotope,  $^9\text{Be}$ .

Because  $^{10}\text{Be}$  connects to aerosols, its residence time in the atmosphere is short (one year) compared to the marine residence time ( $10^2$ - $10^3$  years). However, it remains deposited in rocks, sediments and soil and glacier cores. The residence time of Be inside the soil is about  $10^5$  years, a property that permits the use of  $^{10}\text{Be}$  in geophysics for the dating of evolution processes.

A disadvantage in the use of  $^{10}\text{Be}$  is that this radioactive isotope does not mix enough and in constant proportions with the stable isotope. Therefore, it does not provide a characteristic ratio isotope/element at the moment of its formation. Thus, the stable beryllium cannot be used to correct for dilution or elimination effects as in the case of  $^{14}\text{C}$ .

However, in ocean sediments,  $^{10}\text{Be}$  and  $^9\text{Be}$  almost reach a perfect equilibrium. In other reservoirs, like ice sediments or the earth's atmosphere, the relative  $^{10}\text{Be}$  concentrations can be used to evaluate the variations of the isotope content.

The medium residence time in the atmosphere of about one year makes this radioisotope a good tracer for different intensity variations of cosmic radiations. These variations are the result of solar activity oscillations. Thus, in the high solar activity periods (great number of solar spots), the cosmic radiation screening effect appears due to the intense highly magnetized plasma region produced by the Sun and spread in the cosmic space. In these periods, only low intensity proton fluxes in the atmosphere are registered and, consequently, the production of beryllium has a minimum.

The history of  $^{10}\text{Be}$  is very well preserved in ice deposits from Greenland and Antarctica. From the atmosphere  $^{10}\text{Be}$  is eliminated through rains. Annual ice layers can be distinguished by measuring the ratio  $^{18}\text{O}/^{16}\text{O}$ . This ratio has a biannual oscillation, with a maximum in summer and a minimum in winter.

Generally, the observations proved the existence of an anti-correlation of the  $^{10}\text{Be}$  content in the ice layers of glaciers with the number of the solar spots in the last 400 years. This is only one effect of the AMS usage of  $^{10}\text{Be}$  in understanding and determining the solar activities in the past as well as in the present times.

The  $^{10}\text{Be}$  measurements on the glacier cores can also be used in geomagnetic field history by long time observations (hundreds or thousands of years). Beryllium low mobility once attached on minerals surface lead to the  $^{10}\text{Be}$  use in marine sedimentation rates, in Mn "nodules" growth and in plate glittering in volcanic eruptions. The resistance time deep inside the sea is long enough for the side mixing to make the  $^{10}\text{Be}$  concentration in deep waters almost constant even if some differences were noticed in the deep waters of the oceans.

In the past, erosion rates of rocks, ice stage timing and young volcanoes flowing were areas difficult to explore because an adequate chronometer was missing. In our days, the

AMS technique is used in many soil erosion studies. Also, under special conditions, when production start time is without inherited  $^{10}\text{Be}$  and well defined, then the  $^{10}\text{Be}$  concentration measuring can be used for soil age determination.

AMS also allows the measurement of  $^{10}\text{Be}$  produced directly on the rocks by way of cosmic radiation irradiation on the Earth surface. The production rate of  $^{10}\text{Be}$  depends on the altitude and latitude where the probes were collected. Thus, even at the sea level, the production rates are sufficient for the thousands years exposure to produce enough  $^{10}\text{Be}$  to be measured by AMS. Once the production rates “in situ” are known [6], the sample exposure history can be determined with high precision by measuring only a few isotopes with different life-spans. It is very useful to combine the measurements of several isotopes to ensure the absence of meteoric contaminations and obtain the exact erosion rates. This method was used for meteor crater dating and erosion rate determinations in Antarctica rocks.

$^{10}\text{Be}$  was an important nuclide used in meteorites study even when they were used only as conventional techniques.  $^{10}\text{Be}$  measuring was used for the study of Stone Age and Iron Age meteorites, for studying moon and interplanetary dust samples. In most cases, since there is no significant contribution of solar cosmic rays, the production of  $^{10}\text{Be}$  being due to the galactic cosmic rays, such measurements can be used as indicators of the exposure time [7]. In many cases, the exposure time can offer important information about the object origin. For example, using the determined value of exposure time it was proved to be true that the meteorites from the Mac Alpine Hill in Antarctica originate from the moon [8].

Furthermore, because of its low solubility in non-acid waters the  $^{10}\text{Be}$  measuring was applied in Hydrology.

Even more, recent studies using  $^9\text{Be}$  can be found in [9], [10], [11] and [12]-for geosciences, in [10], [11], [12] and [13]- for polar ice, in [13]- for sediments, in [11]- for soils and in [14] – for meteorites.

### **3.3. $^{26}\text{Al}$ Radio-isotope**

Although aluminium is toxic for the human body, animals and plants, its biochemistry was seldom studied and was rarely fully understood. Accumulated aluminium within the human body can lead to serious and possibly deadly diseases such as: encephalopathy, osteopathy and micro-cystic anaemia. In alimentation, aluminium is further accumulated from natural sources including water, alimentary additives and vessel contamination during the washing process. Individual daily consumption from natural sources varies from 1 mg to 10 mg. In the USA, high level aluminium consumption is owed to alimentary additives used. It was estimated that the American people eat about 20 mg of aluminium every day from chemical additives. In Europe, the aluminium consumption is far more reduced because of a low additive ratio in aliments and far more restrictive regulations.

Aluminium in potable water is also a natural contamination source. In many parts of the world, the aluminium sulphur is used in the treatment of water. The European Community Directive Council has defined the consumption level of aluminium to 50  $\mu\text{g}/\text{l}$  with a maximum level of 0.2 mg /l. The studies lead to a possible connection between the aluminium content of water and Alzheimer disease. Typical aluminium quantities from

food and drink mean less than 1% of the maximum quantity achieved from pharmaceutical products such as antacid, analgesic, antidiarrhoeal and ulcer drugs.

For years, phosphate gels containing aluminium were used for hyperphosphatemia on patients with kidney chronic diseases. Unfortunately, in some paediatric cases, on hyperphosphatemia control, great amounts of aluminium were necessary (100 mg/Kg every day) and in these cases, bone disease symptoms were noticed.

A major problem in aluminium distribution in the biological system study is the omnipresent nature of the element. In practice, there is a great probe contamination. The detected aluminium by chemical analysis in the fragments prepared from biological material could be from the system itself or from the sample contamination before the analysis. Therefore, the adequate use of an isotopic tracer in the biological system is a technique used to solve this problem. It would also eliminate the possibility of later contamination with a tracer.

Aluminium, the third most abundant element in lithosphere (8%), can be found in nature as  $^{27}\text{Al}$  (100 %), with only one long life radionuclide, which is  $^{26}\text{Al}$  ( $T_{1/2} = 0.716 \times 10^6$  years).

AMS needs for the analysis less than 1mg of  $^{27}\text{Al}$ , at a concentration of  $10^{-13}$  for the radioactive tracer  $^{26}\text{Al}$ . Together with the low isotope abundance used and the extremely decreased radiological risk, this method is extremely useful for research in human toxicology. Furthermore, because it does not interfere with the stable isotope  $^{27}\text{Al}$ , AMS using  $^{26}\text{Al}$  as a tracer can be very important in aluminium metabolism studies but also for pharmacology when this tracer is adjusted to pharmacologic products. Conventional mass spectrometry method is not practical because of the  $^{26}\text{Mg}$  interference.

Recently, AMS with  $^{26}\text{Al}$  and opened a new way for biochemical studies [3], [15] of models and aluminium distribution kinetics "in vitro" and "in vivo". Recent studies were developed about aluminium absorption in different organs and penetration of cellular membrane. Detailed studies about aluminium bio-kinetics in metabolism were developed in laboratories located in Germany (Aachen Hospital and Technical University Munich) and in the USA (West Lafayette University).

Further on,  $^{26}\text{Al}$  radioisotope can also be used in Earth corrosion measurements. In the terrestrial shell (lithosphere) the  $^{26}\text{Al}$  isotope is produced by the reaction of negative muons with silicon:  $^{28}\text{Si}(\mu^-, 2n)^{26}\text{Al}$ . The  $\text{SiO}_2$  is a good target, for  $^{26}\text{Al}$  synthesizes on the earth surface and the production yield of  $^{26}\text{Al}$  decreases with the depth. The terrestrial erosion can then be obtained from the AMS measurement of the  $^{26}\text{Al}$  concentration and by comparing this value to the calculated production in relation of the depth in the Earth's crust.

### 3.4. $^{36}\text{Cl}$ Radio-isotope

$^{36}\text{Cl}$  ( $T_{1/2} = 3.02 \times 10^5$  y) is very soluble in water and has low cohesion on solid surface contacts. It is naturally produced in the higher atmospheric layer (high stratosphere) by spallation reactions on  $^{40}\text{Ar}$  induced by secondary neutrons and protons flux from the cosmic radiation ( $^{40}\text{Ar}(n, p4n)^{36}\text{Cl}$  and  $^{40}\text{Ar}(p, 2p3n)^{36}\text{Cl}$ ).

The nuclide takes part in air transfer between stratosphere and troposphere and finally reaches the Earth's surface through rain. Then, it reaches the subterranean water

reservoirs and it is absorbed by the biomass. Therefore, AMS analysis of  $^{36}\text{Cl}$  can be used to measure the age of the water reservoirs, aquifers, evaporation rates for surface waters, source debit, etc.

$^{36}\text{Cl}$  is also produced in nuclear reactors and nuclear explosions and is a good signature of nuclear pollution produced by nuclear power plants (NPP). There are also additional sources of atmospheric  $^{36}\text{Cl}$  concentrations that can contribute to the tropospheric  $^{36}\text{Cl}$  deposition. These sources are: the cosmogenic *in-situ* production of  $^{36}\text{Cl}$  in the lithosphere, the Chernobyl fall-out and the  $^{36}\text{Cl}$  production by nuclear weapon tests. To obtain a good estimate of the produced nuclear pollution at a certain location, one has to take into account all the other sources of  $^{36}\text{C}$  production.

$^{36}\text{Cl}$  was also used in hydrology, in subterranean water dating (time spent from precipitations falling), in water mix detection from different sources, in water path drawing, in the study of net infiltrations in arid regions, for the study of meteorological conditions in the past [16], [28], for evaporating rate [17], etc. However,  $^{36}\text{Cl}$  cannot be used for subterranean waters dating with the same confidence level as  $^{14}\text{C}$ . This is due to  $^{36}\text{Cl}/\text{Cl}$  ratio modifications in water, which is not determined by the disintegration process.

An important contribution to  $^{36}\text{Cl}$  inventory was represented by the bomb explosions in 1953 and 1958. A concentration peak for  $^{36}\text{Cl}$  was observed in the Alps and the Dye-3 glacier during this period of time (the bomb-test peak was approximately 3 orders of magnitude above natural levels) [18, 19]. One can conclude that  $^{36}\text{Cl}$  is an important atmospheric tracer and a good indicator of nuclear pollution.

### 3.5. $^{129}\text{I}$ Radio-isotope

After 1945, the radiation level inside the oceans has clearly been increasing due to nuclear experiments that had been contaminating very large sea regions. An important contribution to sea or ocean contaminations were not only the radioactive discharges of  $^{129}\text{I}$  from nuclear reprocessing plants but also the contaminations from vessels using nuclear propulsion and navigating all over the sea and oceans. During their cruises, considerable quantities of fission products are discharged into the planetary ocean.

$^{129}\text{I}$  ( $T_{1/2}=15.7$  Myr) is a fission product. Starting with the 60's, the  $^{129}\text{I}$  was emitted by nuclear installations either in the liquid or in the gas form and it became the highest source of nuclear pollution for the environment. Furthermore, between 1966 and 1994 the biggest Western Europe nuclear reprocessing plants from La Hague (France) and Sellafield (Great Britain) contributed with an input of 906 kg of  $^{129}\text{I}$ , discarded in the English Channel and an input of 534 kg of  $^{129}\text{I}$ , discarded in the Ireland Sea.

The only stable isotope of iodine is the  $^{127}\text{I}$ . The natural production of the  $^{129}\text{I}$  is based on the spallation reaction of cosmic rays on atmospheric Xe with a low production yield. As a consequence, the natural background for the measurements is practically negligible. Due to its high volatility, when entering the atmosphere, the  $^{129}\text{I}$  is transported over large distances. The major problem in the application of the  $^{129}\text{I}$  has always been the combination of a low natural abundance with the long average life, which results in a very low activity. Given the required limit of detection, which is usually dictated by the mass of available sample (some milligrams of iodine), detection is only possible to achieve by using the AMS technique.

In the light of the ultra-sensitive measurements of the  $^{129}\text{I}$ , the monitoring possibilities of the AMS were evaluated by the Vienna Agency of Atomic Energy (IAEA). This was accomplished as a follow up to the inter-comparison exercise from July 1997, which was done in cooperation with the U.S. Department of Energy and the International Safeguards Division. As a result, IAEA has identified and recommended the measurement of the  $^{129}\text{I}$  as a potential signature of the reactor or reprocessing operations, or for the detection of undeclared nuclear activities [20].

The detection of  $^{129}\text{I}$  using classic measurement techniques is very difficult. AMS is the only method capable to determine the  $^{129}\text{I}$  content with  $10^6$  atoms detection sensitivity and isotopic ratio  $^{129}\text{I}/^{127}\text{I} = 10^{-14}$  [21].

As presented in the introduction of this paper, an important task is the prevention and monitoring of the nuclear pollution of the planetary ocean. AMS with  $^{129}\text{I}$  is a useful tool for such a task. The iodine discharged into the ocean will suffer a strong dispersion due to ocean water currents.

$^{129}\text{I}$  can also be used as a tracer to study hydrodynamic mechanisms inside the oceans. Thus, iodine is present in sea water under two ionic forms:  $\text{IO}_3^-$  and  $\text{I}^-$ . This makes iodine to have a great solubility and water mobility and it can be easily transported over long distances by the ocean water currents.  $^{129}\text{I}$  tracer studies lead the researches to a better knowledge of the oceanic water circulation and deep waters current formation [22, 23, 24, 25, 26].

In case of a nuclear accident, iodine is considered to be one of the most dangerous emitted elements. It is discharged into the atmosphere and from here it spreads into water, vegetation, animals, etc. In such situations, the main emitted isotope is  $^{131}\text{I}$  ( $T_{1/2} = 8$  days). Due to its very short half-life it can be measured only for a short period of time but it has a severe radiologic consequence. The  $^{129}\text{I}$  fraction emitted into the atmosphere is harmless from this point of view. However, since it has a very long half-life it can be followed over a long time.  $^{129}\text{I}$  measurements made after the Chernobyl accident, could be used to trace the iodine evolution in the atmosphere, biosphere, and hydrosphere [27].

In order to determine this fission product's mobility induced by mineral erosion and its migration in mineral waters [28], in hydrology, studies about  $^{129}\text{I}$  were made on uranium minerals. Because of its mobility, iodine can escape from uranium minerals and dissolve in subterranean waters. These types of studies are very interesting because the uranium minerals can be considered in geo-chemistry to be an analogue of nuclear combustible deposits. Thus, useful information about path and velocity prediction of activities around nuclear combustible deposits is very interesting for radioprotection [29].

In geology,  $^{129}\text{I}$ , based on its very long life time and biofilic nature, was used to investigate the petroleum synthesis and migration, about the salted water that follows it [30] and about diagenetic processes of marine sediments [31].  $^{129}\text{I}$  can also be used for dating applications up to  $10^8$  years period of time [32].

#### 4. Conclusions

AMS together with ERDA, RBS, NRA, PIXE, PIGE perform elemental analysis with a manifold application area. When the research requires highly sensitive analysis, detecting the atoms individually, AMS is the only method able to accomplish such a task. It

represents today a complementary experimental method for the other classical nuclear and atomic analyzing methods.

Environmental physics, biophysics, medicine, geology, anthropology, forensic science, etc. are some of the researches to benefit from the high analyzing power of AMS. Whenever human research requires a measurement at the nanoscale of a process, AMS might be applied.

The aim of the present paper was to give an introduction in the AMS analyzing technique and to present some of the possible applications. The paper presentation was meant as a guide for a user from any areas of human activity.

### End Notes:

- [1]. P. E. Damon, "Radiocarbon dating of the shroud of Turin", *Nature*, 611, 337, 1989.
- [2]. Stan-Sion, M. Enachescu, M. S. Dogaru, "Environmental radio nuclides measured by AMS", Advanced Training Course: New Techniques for the Detection of Nuclear and Radioactive, Mugla, Turkey, 2008.
- [3]. J. Baker, J. P. Day, T. W. Aitken, T. R. Charlesworth, R. C. Cunningham, P. V. Drumm, J. S. Lilley, and G. W. A. Newton and J. M. Smithson, "Development of aluminum – 26 accelerator mass spectrometry for biological and toxicological applications", *Nucl. Instr. and Meth. B*, 52, 540-543, 1990.
- [4]. K. Spindler, "The iceman's last week", *Nucl. Instr. and Meth. B*, 92, p 274-281, 1994.
- [5]. W. S. Broecker, T. H. Peng, S. Trumbore, G. Bonani and W. Wolfli, "The distribution of radiocarbon in the glacial ocean", *Biogeochemical Cycles*, 1990.
- [6]. K. Nishiizumi, E. I. Winterer, C. Kohl, J. Klein, R. Middleton, D. Lal and J. R. Arnold, "Cosmic ray production rates of  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in quartz from glacially polished rock", *J. Geophys. Res.* 94, 17907-17915, 1989.
- [7]. K. Nishiizumi, M. Imamura, C. P. Kohl, H. Nagai, K. Kobayashi, K. Yoshida, H. Yamashita, R. C. Reedy, M. Honda and J. R. Arnold, " $^{10}\text{Be}$  profiles in lunar surface rock 68815", *Proc. Lunar Planet. Sci. Conf. 18 th*, pages 79-85, 1987.
- [8]. O. Eugster, M. Burger, U. Krhenbhl. Th. Michel, J. Beer, H. J. Hofmann, H. A. Synal, W. Woelfli and R. C. Finkel, "History of the paired lunar meteorites MAC 88104 and MAC88105 derived from noble gas isotopes, radionuclides, and some chemical abundances", *Geochim. Cosmochim. Acta*, 55, 3139-3148, 1991.
- [9]. G. M. Raisbeck, F. Yiou, D. Bourles, J. Lestringuez and D. Deboffe, "Measurement of  $^{10}\text{Be}$  with tandem accelerator operating at 2 MeV", *Nucl. Instr. and Meth. B*, 5, 175-178, 1984.
- [10]. L. Brown, " $^{10}\text{Be}$ : Recent applications in the Earth science", *Phil. Trans. R. Soc. Lond.*, A323, 75-86, 1987.
- [11]. L. R. McHargue and P. E. Damon, "The Global Beryllium – 10 cycle", *Rev. of Geophys.*, 29, 141-158, 1991.
- [12]. D. L. Bourès, "Beryllium isotopes in the Earth's environment", *Earth Planet. Sci. Lett.*, 96, 287-304, 1990.
- [13]. R. F. Anderson, Y. Lao, W. S. Broecker, S. E. Trumbore, H. J. Hofmann and W. Wolfli, "Beryllium isotopes in the Earth's environment", *Earth Planet. Sci. Lett.*, 96, 287-304, 1990.
- [14]. K. Nishiizumi, " $^{53}\text{Mn}$ ,  $^{26}\text{Al}$ ,  $^{10}\text{Be}$  and  $^{36}\text{Cl}$  in meteorites data compilation", *Nucl. Track Radiations Meas.*, 13, 209-273, 1987.
- [15]. O. Meirav, R. A. L. Sutton, D. Fink, R. Middleton, J. Klein, V. R. Walker, A. Halabe, D. Vetterli and R. R. Johnson, "Application of accelerator mass spectrometry in aluminum metabolism studies", *Nucl. Instr. Meth. Phys. Res.*, 52, 536-539, 1990.
- [16]. M. Paul, A. Kaufman, M. Magaritz, D. Fink, W. Henning, R. Kaim, W. Kutschera and O. Meirav, "A new  $^{36}\text{Cl}$  hydrological model and  $^{36}\text{Cl}$  systematic in the Jordan River/Dead Sea system", *Nature*, A321, 511-515, 1986.

- [17]. M. Margaritz, A. Kaufmann, M. Paul, E. Boaretto and G. Hollos, "A new method to determine regional evapotranspiration", *Water Res.*, 26, 1759-1762, 1990.
- [18]. H.A. Synal, j. Beer, G. Bonani, M. Suter, W. Wolfli, " Atmospheric transport of bomb-produced  $^{36}\text{Cl}$ , *Nucl. Instr. Meth. Phys. B52*, 483-488, 1990.
- [19]. H. A. Synal, "Beschleunigermassenspektrometrie mit  $^{36}\text{Cl}$ ", *Phd, ETH Zurich*, 1989.
- [20]. UCRL-ID-128218, in Rep. LLNL, July, 1997.
- [21]. C. Stan-Sion, O. Constantinescu, M. Enachescu, M. Dogaru, A. Petre, I. Stefanescu, C. Pearsica, "Monitoring the nuclear pollution in the environment by ultra-sensitive measurements of tritium and 129-iodine", *13th Progress in Cryogenics Separation, ICTI, Rm. Valcea, Romania*, November 7-9, 2007.
- [22]. Ch. Heinze, P. Schosser, K. P. Koltermann and J. Meincke, "A trace Study of Deep Water Renewal in the European Polar Seas", *Deep Sea Research*, 37, 1425, 1990.
- [23]. P. Schlosser, G. Bonisch, M. Rhein and R. Bayer, "Reduction of Deepwater Formation in the Greenland Sea during the 1980's". *Evidence from Tracer Data, Science*, 251, 1054, 1991.
- [24]. H. D. Livingston, J. H. Swift and H. G. Ostlund, "Artificial Radionuclides Tracer Supply to the Denmark Strait Overflow between 1972 and 1981", *Journal of Geophysical Research*, C4, 6971, 1985.
- [25]. K. Aagaard, J. H. Swift and E. C. Carmack, "Thermohaline Circulation in the Artic Seas", *Journal of Geophysical Research*, C3(90), 4833, 1985.
- [26]. A. J. Watson, R. R. Dickinson, E. M. Gmitrowicz, "Deep-water Renewalin the Northern North Atlantic", *Nature* 344, 848, 1990.
- [27]. M. Paul, D. Fink, G. Hollos, A. Kaufman, W. Kutchera and M. Magaritz, "Measurement of  $^{129}\text{I}$  Concentrations in the Environment after the Cernobil Reactor Accident", *Nucl. Instr and Meth B*, 29, 341, 1987.
- [28]. J. Fabrika-Martin, S. N. Davis and D. Elmore, "Applications of  $^{129}\text{I}$  and  $^{36}\text{Cl}$  in hydrology", *Nucl. Instr. and Meth. B*, 29, 361-371, 1987.
- [29]. J. Fabrika-Martin, S. N. Davis, D. Roman, P. L. Airey, D. Elmore and P. W. Kubick, "Iodine -129 and Chloride -36 in Uranium Ores", *Chemical Geology*, 72, 7, 1988.
- [30]. U. Fehn, S. Tullai-Fitzpatrick, R. Teng, H. E. Gore, P. Sharma and D. Elmore, "Dating of oilfield brines using  $^{129}\text{I}$ ", *Nucl. Instr. And Meth B*, 52, 446, 1990.
- [31]. Fehn, R. Holder, D. Elmore, T. Brunelle, R. Teng and P. W. Kubick, "Determination of natural and anthropogenic I in marine sediments", *Geophysical Research Letters*, 13, 137, 1986.
- [32]. Fabryka Martin, H. W. Bentley and D. Elmore and P. L. Airey, "Natural  $^{129}\text{I}$  as an enviromental tracer", *Geochimica et Cosmochimica Acta*, 429, 337, 1988.