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Based on Nuclear and Atomic Physics
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Report WP10 IDRANAP 81-04/2004

AMS WITH ^{129}I FOR PREVENTING AND MONITORING ACCIDENTAL OR DELIBERATE DISCHARGE OF SMALL NUCLEAR DEBRIS*

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* Work supported in part by the Center of Excellence IDRANAP under Contract with European Commission ICA1-CT-2000-70023, and by Ministry of Education and Research of Romania

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1. Introduction

Accelerator Mass Spectrometry (AMS) is a relatively new detection technique, which constitutes today the highest sensitive method for counting individual radioactive atoms with detection sensitivity 1 atom in 1 million of billions of surrounding atoms (10^{-15}).

The extremely high detection sensitivity of AMS in conjunction with the enhanced values observed for ^{129}I relative to other fission products makes the measurement of ^{129}I with AMS an efficient tool for nuclear safeguards, detecting and preventing accidental or deliberate discharge of small nuclear debris that otherwise would remain undetected by radioactivity measurements. Such measurements are applied all over the world however in Eastern Europe there is no facility doing such measurements for a large geographical area.

The goal of the project is to measure, monitor and investigate the transport of ^{129}I in vicinity of three nuclear power plants in Eastern Europe: Kosloduy (Bulgaria), Chernavoda (Romania), Chernobyl (Ukraine).

In order to achieve this task a new high-resolution detection system has been constructed at the AMS facility in Bucharest, which came recently in operation. The detection system is a gas-semiconductor (E-multi-ΔE) system with Time of Flight (TOF) discrimination, providing a good isobaric separation. It consists of two symmetric anodes plates, providing the ΔE signals, a Frisch grid and the cathode. The TOF base is 3.5 m long. A micro-channel plate detector and the Si-surface barrier detector give the start/stop signals, respectively. The detector system works in coincidence with the TOF system. The associated electronics contains 4 preamplifiers (PA), 3 Spectroscopic Amplifiers (SA), 3 Timing filter amplifiers (TFA), 3 constant fraction timing discriminators (CFTD), 1 time to amplitude convertor (TAC), 4 HV bias - power supplies.

2. Natural and anthropogenic inventory of ^{129}I in the environment.

The half-life of ^{129}I is extremely long, 15.7 Ma, however, still to short in comparison to the age of nucleo-synthesis estimated at c.a. 10Ga to be considered a primordial radionuclide. Its beta minus disintegration goes down to ^{129}Xe.

The atmospheric production of ^{129}I is due to the spallation reactions induced by the cosmogenic radiation on atmospheric Xe and due to spontaneous fission of Uranium from the earth's crust or from the water of oceans.

The estimated concentration of ^{129}I in oceans is 3.7 E-4 mol/m^3, corresponding to an isotopic concentration ^{129}I/^{127}I = 0.25E-12. According to our estimates the atmospheric production brings only 1% contribution to the total inventory. The lowest concentrations were measured to be 0.4 E-12 in the North Polar Sea. However, in rivers and laces the iodine isotopic concentration is a factor 10^2 to 10^3 higher.

The anthropogenic contribution to the ^{129}I inventory is due expelled material from Nuclear Power plants and from nuclear bomb tests. In Nuclear Power plants the ^{129}I is produced from fission of ^{235}U and ^{238}U and of the breaded ^{239}Pu with fission yields of 0.83%, 1.65% and 1.53%, respectively. Very important are the inputs produced by the releases of nuclear fuel reprocessing plants as La Hague (F), Sellafield (GB) and West Valley (SUA). The released ^{129}I is for the two locations in Europe ca. 2360 kg until 1997 and only 290kg in the USA.

Adding these contributions to the inventory in Ocean water from the Northern hemisphere and assuming uniform mixing the isotopic ratio $^{129}I/^{127}I$ is ca. 4E-9. This value is in good resemblance with values measured at different locations, as listed in table 1.

Table 1. 129I concentrations measured in rainwater samples collected at locations in Europe and USA.

Location	$^{129}I/^{127}I$	Ref.
Uppsala, Sweden	$(2.6\pm0.2)\cdot10^{-7}$	[1]
Westerland, Germany	$(7.4\pm3.0)\cdot10^{-7}$	[2]
Mailand, Italia	$(2.0\pm0.2)\cdot10^{-7}$	[1]
Indiana, USA	$(8.3\pm0.3)\cdot10^{-9}$	[3]
Texas, USA	$(1.7\pm0.2)\cdot10^{-9}$	[3]

For comparison, in table 2, values from locations near to the nuclear fuel reprocessing plants are given. These concentrations are about 2 times larger as the averaged measured values.

Table 2. Iodine 129 at locations close to nuclear fuel reprocessing plants.

Location	$^{129}I/^{127}I$	Ref.
Seetang-La Hague(1991)	$(1.5\pm0.2)\cdot10^{-6}$	[4]
Heysham–Sellafield (1992)	$(8.9\pm1.2)\cdot10^{-7}$	[4]
West Valley (1994)	$(7.2\ )\cdot10^{-5}$	[5]
Rainwater Munich (1986)	$(3.9)\cdot10^{-6}$	[6]

The goal of our WP was to establish a monitoring procedure based on measurements of ^{129}I with AMS, in order to detect and prevent accidental or deliberate discharge of small nuclear debris from nuclear power plants(NPP).

Water samples were collected from the Danube River in the vicinity of the two nuclear electric power plants in operation at **Kosloduy-Bulgaria (VVER-system) and Cernavoda – RO (Candu-system).** Water samples were collected also from the Black Sea .

Fig. 1 presents the geographic map of the Danube River flow (lower path) through Serbia, Bulgaria and Romania.

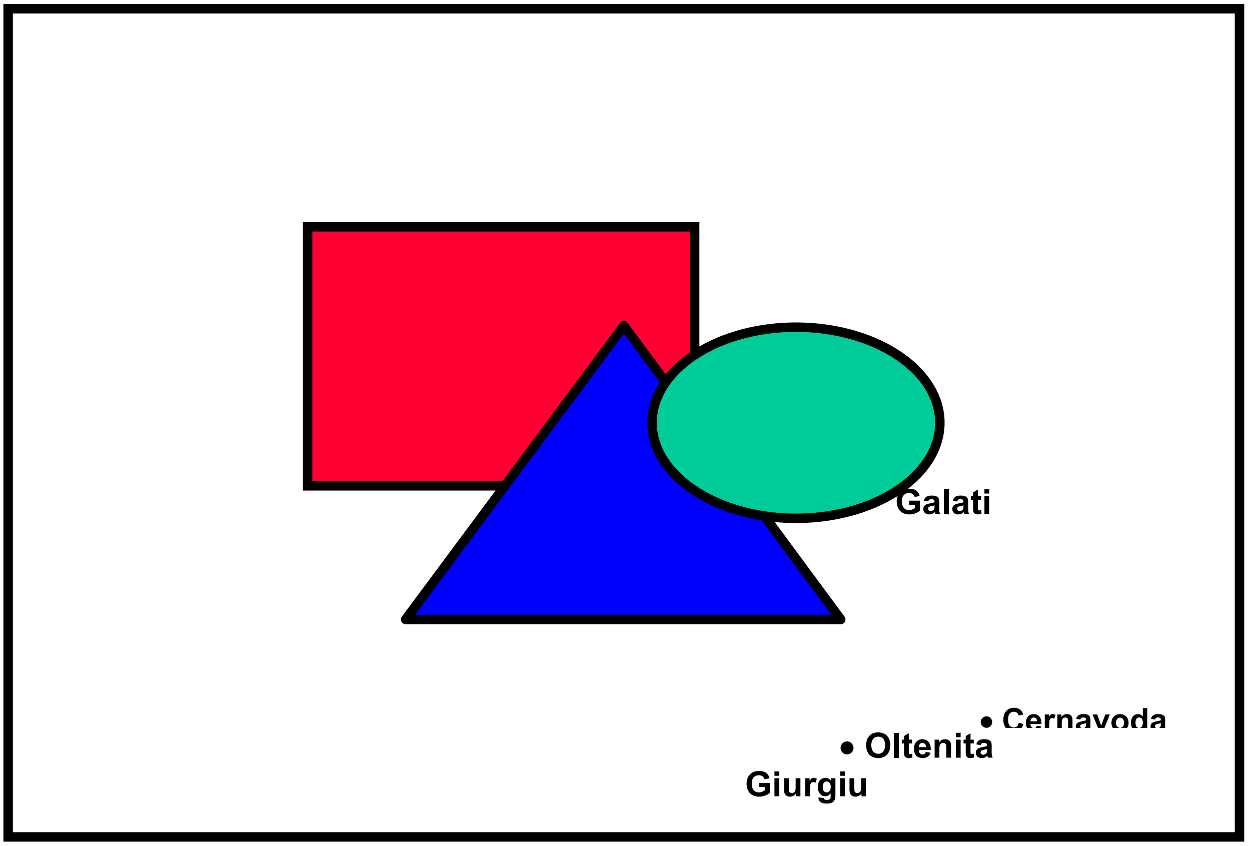

Fig. 1. The Danube River on its lower flow path up to the ending through its Delta in the Black Sea.

The Kozloduy Nuclear Power Plant is situated in Bulgaria on the Danube River, exactly opposite site to the Romanian town Giurgiu. The six groups of the nuclear power plant utilise water from the Danube river for their cooling systems.

Water samples for 129I-AMS measurements at this Nuclear Power Plant were collected from the Danube River at Giurgiu, opposite to Kozloduy (index 3B), and from Oltenita (index 4B) located some kilometres downstream.

The Nuclear electric power plant from Cernavoda-Romania is of type CANDU – PHWR-6 (Canadian Deuterium Uranium – Pressurised Heavy Water Reactor). In the late 1970s a five-unit nuclear power plant was planned at Cernavoda, on the Danube River. After considering carefully both Russian VVER-440 and Canadian CANDU technology it was decided to adopt the latter. Cernavoda was based on technology transfer from Canada (AECL), Italy and the USA, with Candu-6 heavy-water reactors.

Construction of the first unit started in 1980, and of units 2-5 in 1982. In 1991 work on the latter four was suspended in order to focus on unit 1, responsibility for which was handed to an AECL-Ansaldo (Canadian-Italian) consortium. The state nuclear power corporation Societatea Nationala Nuclearelectrica (SNN), established in 1998, operates Cernavoda1 (655 Mwe) and is now completing unit 2.

Cernavoda-1 has been using 105 tonnes of natural uranium oxide fuel per year, which is fabricated by the SNN subsidiary Pitesti Fuel Plant. At the end of 2003 it started making slightly enriched uranium fuel, and in preparation for unit 2 commissioning, its capacity is being doubled.

Heavy water is produced in the south-west of the country, near Drobeta-Turnu Severin. Spent fuel is stored at the reactors for up to ten years. A dry storage facility for spent fuel is being built, based on the Macstor system designed by AECL. The first module was commissioned in 2003. Preliminary investigations are under way regarding a deep geological repository. Near Cernavoda, a low- and intermediate-level waste repository is envisaged from 2005. A radioactive waste treatment facility operates at Pitesti.

The cooling system consists at each unit of two closed circuits using as coolant heavy water and an open circuit, using the water from the Danube River. The normal, water is taken form the Danube channel and after using it to covert the thermal energy to electric energy it will be then released again to the Danube River. Water samples for AMS analyse were collected form locations at the in- and outlet of the cooling loop (positions 12-index 2B and 13-index 1B) .

Fig.2 presents the location of Cernavoda Nuclear Power Plant and the water sample collection points at the in- and outlet of the cooling loop (positions12 and 13) .

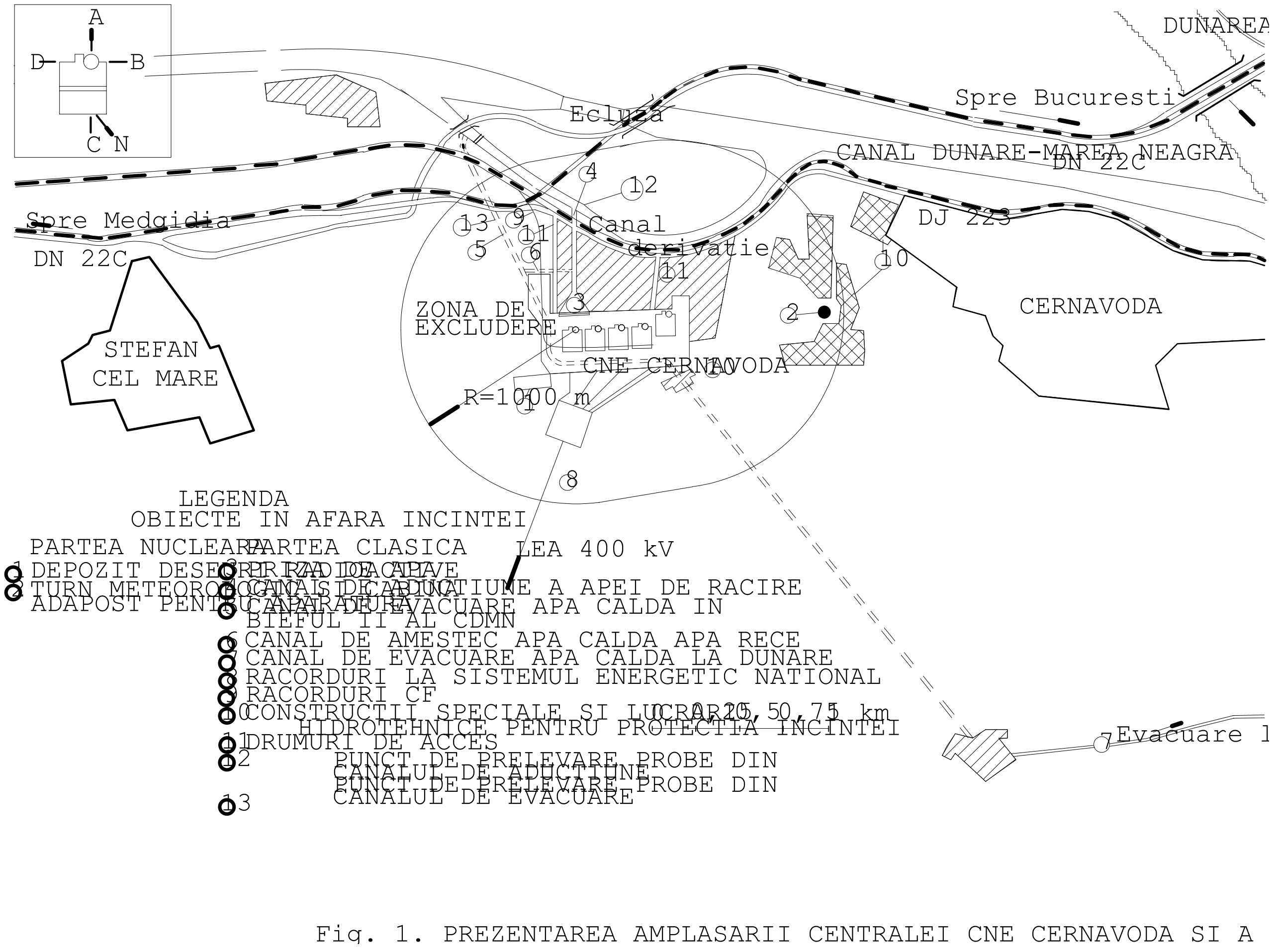

Fig. 2 . General lay out of Ceranvoda Nuclear Power Plant on the Danube River.

Finally, our ^{129}I AMS measurements were performed also on water samples collected downstream from the Nuclear Power Plant, at Galati - a big harbor at the Danube River, before the Danube Delta ending in the Black Sea (Sample index 5B). Moreover, water of the Black Sea was also measured. Sampling was performed at Mangalia –harbor (Sample index 6B). At each location 3 up to 5 water samples were collected and analyzed by AMS. In table 3, resuming the experimental results only averaged values are presented for each location.

Results and Discussion.

The General layout of the AMS facility in Bucharest is shown in fig.3, below. Details concerning the facility and detection systems can be found in IDRANAP Report 15_01/2001, 16_01/2001.

A bi-parametric AMS spectra (TOF, Energy) from measuring 129I is shown in fig.4.

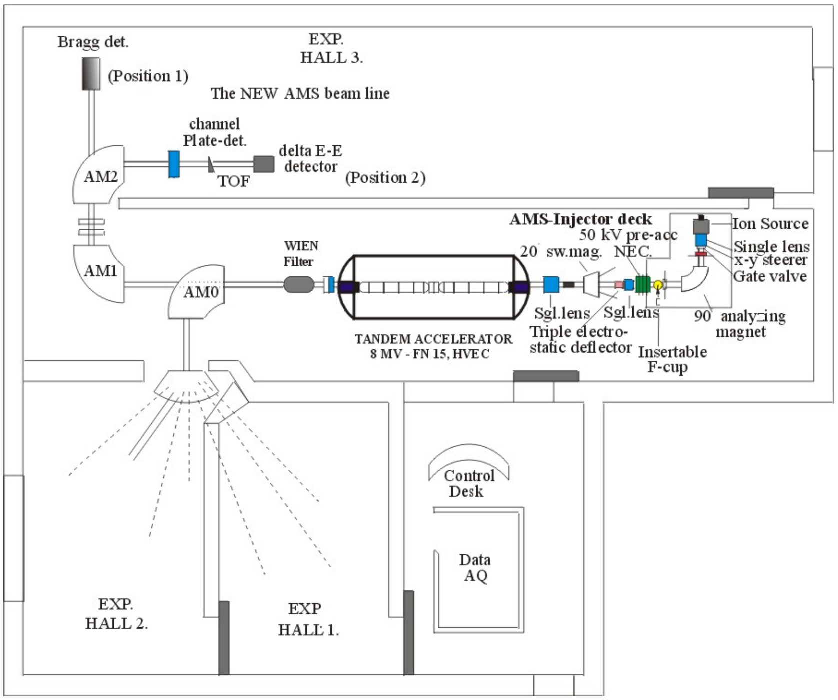

Fig.3 General lay out of the AMS facility in Bucharest.

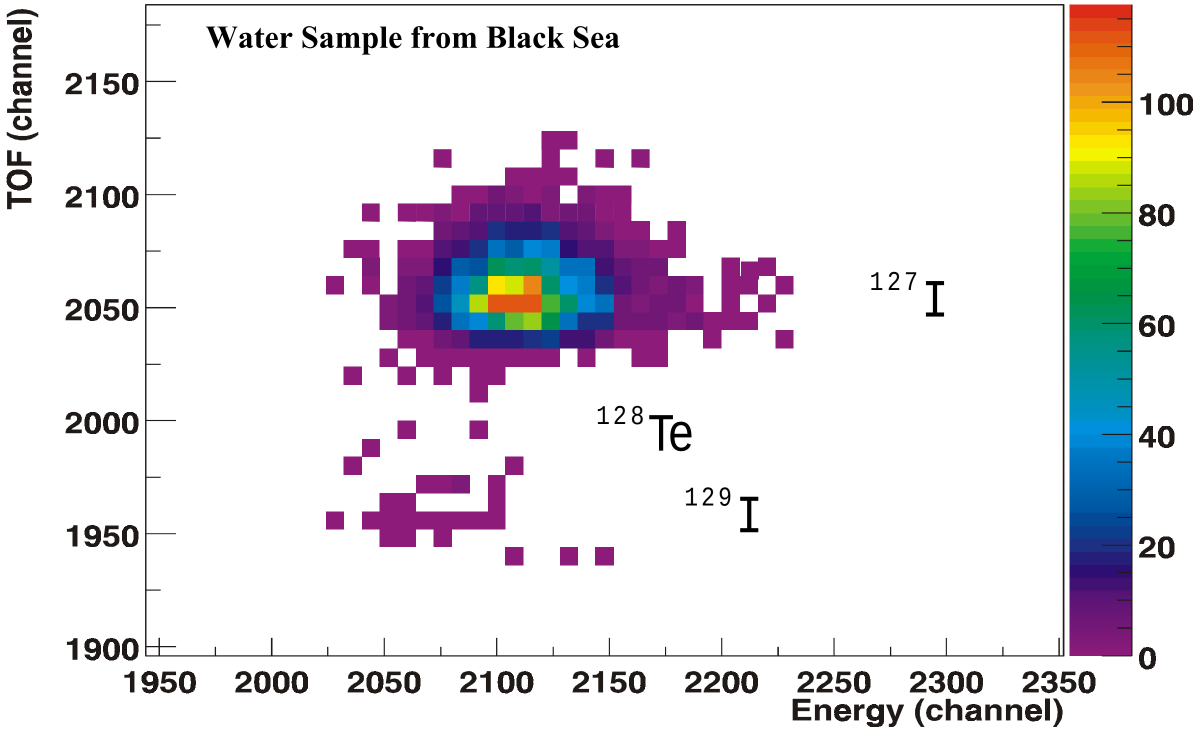

Fig.4 Bi-parametric TOF-E spectra measured by AMS.

The obtained AMS results are presented in Table 3.

The ^{127}I concentration in water samples was determined by spectrophotometry.

At the date of the sample collection , July 12, 2003 , no notable differences of ^{129}I concentration could be observed in the different locations on the Danube River. The averaged concentration value for ^{129}I in the Danube River was (6.4 ±1.2)x107 atoms/l. This corresponds to iodine concentrations observed in many European lakes (see Table 4).

Table 3. AMS measurements of ^{129}I concentrations in the Danube River and Black Sea.

Index	Location	$^{129}I / ^{127}I$*	^{129}I (atoms/l)
1B	Cernavoda 1	$(5.1\pm1)\ 10^{-12}$	$(7.0\pm1.4)\ 10^{7}$
2B	Cernavoda 2	$(3.6\pm0.5)\ 10^{-12}$	$(4.9\pm0.7)\ 10^{7}$
3B	Giurgiu	$(5.4\pm0.4)\ 10^{-12}$	$(7.4\pm0.5)\ 10^{7}$
4B	Oltenita	$(4.9\pm1)\ 10^{-12}$	$(6.7\pm1.4)\ 10^{7}$
5B	Galati	$(3.0\pm1.8)\ 10^{-12}$	$(4.1\pm2.5)\ 10^{7}$
6B	Black Sea	$(3.3\pm0.3)\ 10^{-12}$	$(8.6\pm2.9)\ 10^{7}$

* Blank corrected values

Table 4. ^{129}I concentrations in lakes.

Location	$^{127}I/^{129}I$	^{129}I (atoms/l)
Ammersee	$(1.0\pm0.3)\ 10^{-12}$	$(3.9\pm1.2)\ 10^{7}$
Lago Maggiore	$(6.2\pm4.0)\ 10^{-13}$	$(1.9\pm1.2)\ 10^{7}$
Gardasee	$(2.2\pm0.4)\ 10^{-12}$	$(6.8\pm1.2)\ 10^{7}$
Baikal lake	$(5.6\pm1.0)\ 10^{-13}$	$(9.9\pm1.8)\ 10^{6}$

For the Balck Sea the concentrations was a little higher, $(8.6\pm2.5) \times 10^{7}$ atoms/l. Thus, not indication exists for any radioactive contamination introduced on the Danube River or Neper River (biggest rivers ending in the Balck Sea).

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