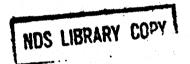


INTERNATIONAL NUCLEAR DATA COMMITTEE

PROGRESS REPORT

ON NUCLEAR DATA ACTIVITIES IN ROMANIA



Compiled by

S.N. Rapeanu

February 1988

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FOREWORD

This progress report contains the main nuclear data work performed during the year 1987 in the institutes of the Central Institute of Physics from Romania. It has been prepared to promote exchange of nuclear data information between the Socialist Republic of Romania and the other member states of IAEA. The emphasis in the works here reported has been on calculations, measurements and evaluations of nuclear data for application. The individual reports are not intented to be complete or formal. Consequently they should not be quoted and reproduced without the permission of the authors.

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G.D.H. pre-equilibrium emission model and statistical model parameters for structural material fast neutron data calculations*

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Inclusion of the angular momentum conservation in the Geometry Dependent Hybrid (GDH) pre-equilibrium emission subroutine of the Hauser-Feshbach code STAPRE is discussed. The consistency of the statistical model nuclear level density and the equivalent particle-hole state density has been achieved following a unitary use of an energy dependent level density parameter.

The neutron and charge particles optical model potentials selected from literature are commented. The El gamma-ray strength functions, used in the gamma-ray transmission coefficient evaluation, have been taken from an empirically modified energy-dependent Breit-Wigner (EDBW) model.

The paper account of the nuclear level density over a large energy range has been obtained through the use of the empirical back-shifted Fermi gas (BSFG) model, at medium excitation energies, and of a realistic analytical formula with microscopic suggested parameters at the high excitation energies. The necessary transition excitation range between the two different density approaches, in the mass range 40 < A < 65, has been discussed.

^{*}A paper on this subject was presented at the IAEA First Research Coordination Meeting on Methods for the Calculation of Fast Neutron Nuclear Data for Structural Materials (Bologna, 7-10 October 1986), as progress report for the research contract 3802/R1/RB, with the above abstract.

Three groups of U natural decay modes

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234 U is one of the nuclides for which not only a-decay but also cold fission had been experimentally determined. It was shown (see for example [1]) that ¹⁰⁰ Zr is the most probable light cold fission fragment, corresponding to the heavy fragment ¹³⁴ Te₈₂ with a magic number of neutrons.

Calculations [2-5] within analytical superasymmetric fission model (ASAFM) are in good agreement with experimental results: the half-life for a-decay is accurately reproduced and the preferred cold fission split obtianed in the calculations, 100 Zr + 134 Te, is the same as in experiments.

A third group of decay modes by charged particle spontaneous emission was predicted [2-5] to be ^{24,26} Ne and ²⁸ Mg, radio-activities, having comparable emission rates. Recently, this group was confirmed in a very interesting experiment [6] in which for the first time was detected ²⁸ Mg radioactivity and also for the first time more than one heavy ion radioactivities.

In this way ²³⁴U is the first, and up to now the unique example, of a nucleus for which all three groups of decay modes are measured. It shows that ASAFM is able to describe in a unified way various decay modes in a wide range of mass asymmetry.

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The induced fission probability calculation for Pu

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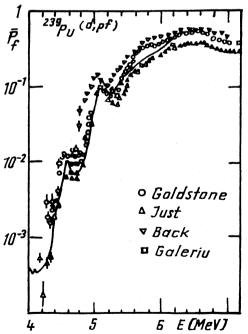
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Using the Strutinsky method, the level densities for all interesting deformations and the parameters of double humped fission barrier for ²⁴⁰Pu were generated in the paper /1/.

A very good agreement between theory and experiment for the neutron interaction cross sections of 239 Pu in the energy range 0.1-1.0 MeV /2/ was obtained, by using these parameters.

This agreement is not enough to check the fission barrier parameters validity, due to known difficulties to reproduce, using the same parameters, the induced fission probability, for a large energy region, because of the resonant structure of the fission probability.

In this paper, the fission probability in ²³⁹Pu (d,pf) reaction, is calculated using the JWKB method in the weak coupling approximation and taking into account both the width level fluctuation and the contributions of the transition states from continuum.



The theoretical result correctly (solid curve - Fig.) describes the experimental data in the underbarrier region.

The good agreement, for the first time, both of the fission probability and the neutron cross-sections /2/, shows on the one hand the validity of the calculated parameters, and, on the other hand, the importance and the necessity of the microscopic selfconsistent calculations in the neutron cross section evaluation.

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Search for pion emission in the thermal neutron induced fission of 235U, and in spontaneous fission of 252Cf

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In paper /1/ it was predicted the possibility of pion emission in processes of fission of heavy nuclei. In order to check this prediction, an experiment was undertaken with nuclear emulsions at the VVR-S reactor in Bucharest. A thick "U target" with 2.43 10²¹ nuclei/cm² was irradiated in a thermal neutron beam. The target was surrounded by nuclear emulsions, 200 thick, in a 4% geometry. The neutron flux was determined by the aid of a 2% pulse ionization chamber in which a 1 µg/cm², high isotopic purity "U target" was placed. Each irradiation was performed in a time integrated thermal neutron flux of 2 10⁸ neutrons/cm².

The experiment with 252 Cf was performed by using a Cf source giving 900 spontaneous fissions/sec. The emulsions were irradiated in a 2π geometry.

The search was conducted for 1-5 prong stors produced by π^- mesons and $\pi^-\mu$ decay of π^+ mesons. Only background events were found. The following upper limits for the Γ_π/Γ_f ratio were established:

Target	Pion energy range	$\frac{\Gamma_{\pi}/\Gamma_{f}}{<6.9 \cdot 10^{-9}}$		
235 _U	8-25 Mev			
252 Cf	8-7.6 Mev	<5.5 10 ⁻⁷		

^{*}Obtained by the kind support of IAEA-Vienna

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Two-photon matrix elements for ns \leftrightarrow n's, n'd transitions in a Coulomb field

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Recently it has been shown [1] that the Kramers-Heisenberg (K.H.) matrix element for any bound-bound two photon transitions may be expressed as a sum of two terms - each involving only one hypergeometric Gauss function ${}_2F_1$.

Using this general method we obtained explicit expressions of the matrix elements for all the two-photon transitions between $ns \longrightarrow n$'s and $ns \longrightarrow n$ 'd states for arbitrary n, n', which give the Rayleigh and Raman scattering cross-sections and the absorption f emission transition rates.

As an example we give the expressions of the invariant amplitudes of the 1s \longrightarrow 4s, 4d and 2s \longrightarrow 3s, 3d transitions:

$$P^{18,48} = \frac{384}{9375} \cdot \frac{1}{\tau(2-\tau)(3-\tau)(4+\tau)^{2}} \cdot \left[\frac{4(\tau-1)^{3}(23\tau^{4}+288\tau^{2}+768)}{(1+\tau)(4+\tau)(5-\tau)} \cdot \right]$$

•
$$_{2}F_{1}(1,-1-\tau,6-\tau;\xi) + 28\tau^{5} - 54\tau^{4} - 258\tau^{3} + 648\tau^{2} - 480\tau + 128$$

$$P^{18,4d} = \frac{2048}{46875\sqrt{10}} \cdot \frac{\tau}{(2-\tau)(3-\tau)(4+\tau)^{2}} \cdot \frac{\left[2(\tau-1)^{3}(7\tau^{2}-48)(7\tau^{2}-48$$

$$p^{28,38} = \frac{32\sqrt{6}}{3125} \left[\frac{18(\tau-2)^{2}(\tau-3)(7\tau^{2}-27)}{\tau(\tau+2)(\tau+3)^{2}(\tau-4)(5-\tau)} \cdot {}_{2}F_{1}(1,-1-\tau;6-\tau;\xi) + \frac{3(\tau^{2}-8\tau+4)(7\tau^{2}-27)}{\tau(\tau+2)(\tau+3)(\tau-4)} + 9 \right]$$

$$P^{28,3d} = \frac{2304}{15625\sqrt{15}} \cdot \frac{\tau}{(\tau+2)(\tau+3)(\tau-4)} \cdot \left[\frac{6(\tau-2)^{2}(\tau-3)}{(\tau+3)(5-\tau)} \cdot _{2}F_{1}(1;-1-\tau;6-\tau;\xi) + \tau^{2} - 8\tau + 4 \right]$$

where $\tau = Z(-2\Omega)^{-\frac{1}{2}}$; $\Omega_j = E_{initial} \pm \omega_j$ (j = 1, 2) are the components of the photon's polarization vectors, Ω_j their energies, the +/- signs in Ω_j being for absorbed/emitted photons; $\xi = \frac{\tau - n}{\tau + n}$ and $F_{\star}(a,b,c;z)$ is the hypergeometric Gauss function.

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Direct radiative recomination cross-sections for arbitrary nS, nP and nD subshells

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Physics Department, Bucharest University

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Exact analytical cross-sections for the direct radiative recombination (DRR) on nS, nP and nD states, with arbitrary principal quantum number n, are obtained in the nonrelativistic dipole approximation. The analytical cross-sections are expressed in terms of elementary functions.

The calculations are made using a method which consists in expressing the wave function of an arbitrary final state (nlm) by acting with a derivative-operator on the wave function of the ground state [1]. This gives the possibility to obtain the cross-section for given l and m and wrbitrary n state by starting with the result for the lowest n, i.e; for n = l+1.

The analytical coross-sections may be used in the case of DFR on Rydberg states of N-electrons ions by considering the ionic charge $Z_{eff} = Z-N$ instead of the atomic charge Z_{eff} .

Numerical values for the DRR cross-sections on nS, nP and nD final states wave obtained for values of principal quantum number n up to 100, in a large range of incoming electron energies involved by the nuclear fusion plasma. These values are in a good agreement with the best relativistic data available [2]. Numerical results compared with the corrected Kramers cross-section [3]

prove that the radiative recombination on the nS, nP and nD states gives at least 96% of the radiative recombination on all the states of the fully ionized atom. It is also proven that the cross-section decreases slowly enough versus n. Indeed for $n \geq 15$ in order that the cross-section decreases by an order of magnitude it is necessary that the principal quantum number n increases at least by 25 units.

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Integral cross sections of (n,p)+(n,n'p) reactions induced by 14.8 MeV neutrons for 95 Mo and 96 Mo

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The natural Mo targets have been exposed at 14.8 MeV neutrons, obtained by the d(T,n) reaction from the neutron generator (type TEXAS-9900). The activation foil exposure have been simultaneously performed with that of the fission chamber containing This fission chamber was absolutely calibrated. The reference cross section is 1.190 b, for $^{238}U(n,f)$ reaction.

The gamma activities have been absolutely measured by the high resolution spectrometry, using a Ge(Li) crystal (100 cm³), absolutely calibrated in efficiency.

The used nuclear data are gathered in the next table:

1.10 2004					
Reaction	$T_{\frac{1}{2}}$	Energy (keV)	Intensi- ty (%)	Abundance (%)	Refe- rence
95 Mo(n,p) 95 Nb 96 Mo(n,n'p) 95 Nb	34.97 days	765.8	99.82	15.9 16.7	2
${}^{95}Mo(n,p)^{95m}Nb$ ${}^{96}Mo(n,n'p)^{95m}Nb$	86.6 hours	235.69	26.1	15.9 16.7	3
96Mo(n,p)96Nb	23.35 hours	568.99 1091.30	57.5 51.02	16.7	2

The cross section values obtained at 14.8 MeV (± 300 KeV), with the associated inaccuracies are given in the second table. In this table are also presented some values obtained in another laboratories, for comparison.

Reaction	Measured cross-section (mb)	(another laboratories) (mb)
95Mo(n,p)95Nb 96Mo(n,n'p)95Nb	35.7±3.2	41.1±3.6 (4) 31.0±4
96Mo(n,p)96Nb 97Mo(n,n'p)97Nb	20.1±1.2	20.8±2.1 19± 1 ⁽⁵⁾ 19± 2 ⁽⁶⁾
96 Mo(n, n'p) 95 m Nb 95 Mo(n, p) 95 m Nb	7.36±0.74	

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Frequency spectra on ZrH U y by inelastic neutron scattering

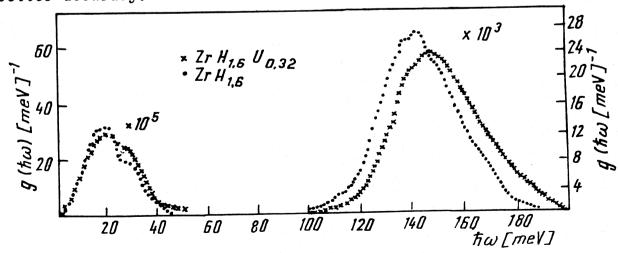
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The dual core test reactor, Pitesti, consists of two TRIGA reactors in one large tank. The 14 Mw Steady State Reactor (SSR) uses as fuel $Zr_{1,6}U_{0,10}$ (\$\frac{235}{4}U\$ under 93% enrichment), the TRIGA-HEU fuel.

In early 1976, General Atomic undertook the development of fuels containing up to 45 wt% uranium in order to allow the use of low enriched uranium LEU), under 20% enrichment, to replace the highly enriched fuels while maintaining long core life. Such researches were undertook within our Institute in the last years in order to produce LEU fuel for TRIGA reactor. For the calculations of neutron flux in the reactor core with LEU fuel the nuclear data were required. In these conditions we performed inelastic neutron scattering measurements using a time-of-flight spectrometer, resolution $\approx 3\%$ and 10^{16} n.cm⁻²/puls on $2\text{TH}_{1,6}\text{U}_{0.32}$ and $2\text{TH}_{1.84}\text{U}_{0.32}$ samples at room temperature.

The obtained frequency spectra on ZrH (moderator) /1/ are compared with the one obtained ZrH U (LEU fuel), presented in the Figure. As one can observe the structure under the both peaks are not rich as under the ones for the moderator and the optic peak only is displaced to the higher energies with \approx 6 meV.

We believe that the data under regarding frequency spectra will be successfully used for the neutron flux calculations with better accuracy.



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On the autocorrelations and frequency spectra in liquid water

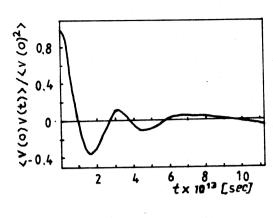
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In the last years, a lot of experimental and theoretical studies have been devoted to the understanding of the structure and dynamical properties of the liquid water. This large interest is explained by several reasons. Now it is already accepted by all scientific investigators that water plays an important role in chemical, biological processes and as moderator or cooling agent in nuclear reactors. Also, water displays a large range of unusual properties /1/, both as pure water and as a solvent. So far, much progress was obtained by using new experimental and theoretical techniques. It is already recognized fact that a complete understanding of the mechanism of the water molecule dynamics required very accurate experimental data.

In the paper /2/, a potential of 2D type is used to obtained the frequency spectrum of the molecular centers from the velocity autocorrelation function. The latter one is calculated from a generalization of the results obtained in the three-pole approximation.

Taking into account mentioned potential and the radial distribution functions /3/ using a model for molecule motion as a whole, we obtained the velocity autocorrelation function and its frequency spectrum for the molecular centers. The spectral function on the proton rotation is separately studied starting from the angular velocity autocorrelations.

Using the theoretical concepts and experimental data from the literature, our experimental data from neutron diffraction /3/ and inelastic neutron scattering data /2/ some parameters were derived /2/. In Fig. 1 and in Fig. 2 are presented velocity autocorrelation function of the water molecules and its frequency spectrum, respectively. The total frequency spectrum obtained as a sum of the translational and rotational bond spectra show the general features of the experimental determined one.



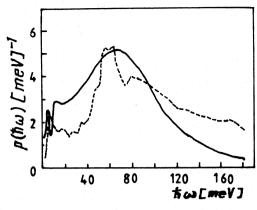


Figure 1

Figure 2

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In-air PIXE analysis using nominal regime protons at Cyclotron

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To determine impurities concentration in liquid and large solid samples in-air PIXE analysis is very advantageous. So, far wear particles determination in engine oils (Fe, Cr, Mn, Cu, Zn, Pb) 3-4 MeV proton beams can be produced from the 6.5 MeV easy-obtained at the U-120 INPE Cyclotron nominal regime protons, extracted through a pressure-air cooled 100 µm aluminium foil into the air, striking the sample after 3-10 cm. To avoid the strong straggling effects large size samples are used († = 50-80 mm).

X-rays are detected through reflection using a 4.3 mm active area diameter horizontal Si(Li) detector (1 mm plexiglas absorber). To minimize the strong gamma-ray background a lead collimator from the extraction foil to the target with helium-gas flowage is used.

Maximum sensitivities have been obtained in the 22 \leq Z \leq \leq 30 region (down to 1 ppm for: Fe and Cu at the Q = 40 nA \times \times 600 s = 24 μ C).

The generalized nonuniform picket-fence model

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The nonuniform picket-fence model for even-even actinides /1/ was generalized for any kind of nucleus (with the spin $I \neq 0$ or I = 0), to calculate the truncated levels contributions for thermal and resonance cross-sections.

The truncated levels are those from outside of resolved resonance range: unresolved resonances and bound levels.

The generalized model takes into account the "s" and "p" wave levels.

The distance between two successive truncated levels is computed based on the average distance for unresolved resonances, the level density parameter and excitation energy of compound nucleus.

The unresolved resonance parameters which are input data, could be dependent on energy.

The generalized model takes into account for the marginal resonance contribution (at the limits of the resolved resonance range) calculated by Breit Wigner Multilevels formalism, as well as for the distant resonances.

The level density parameter is computed by Lynn formalism.

First time, these contributions were calculated by a uniform picket-fence model, developed by Olsen, de Saussure and Perez
/2/.

The generalized model was applied for 18 transactinium nuclei having odd mass numbers using the program RESIN-1.

The analysis of the results shows that, for thermal energies, the contributinos of the truncated levels are equal either with negative resonance contribution, or with the background corrections, for missing negative resonances.

In the resolved resonance range, the contributions are significantly (~40%) for the first part of the energy region. Consequently, the cross section evaluated by adding the corrections given by truncated levels to the Breit-Wigner Single Levels calculations, is improved, especially in the valley between resonances.

In the figures 1 and 2, the dependence of the truncated

level contributions with mass number and energy, for total and fission cross sections is represented.

The general conclusion is that the contributions of the truncated levels could replace either the negative resonance corrections, or background corrections, in the evaluated data files.

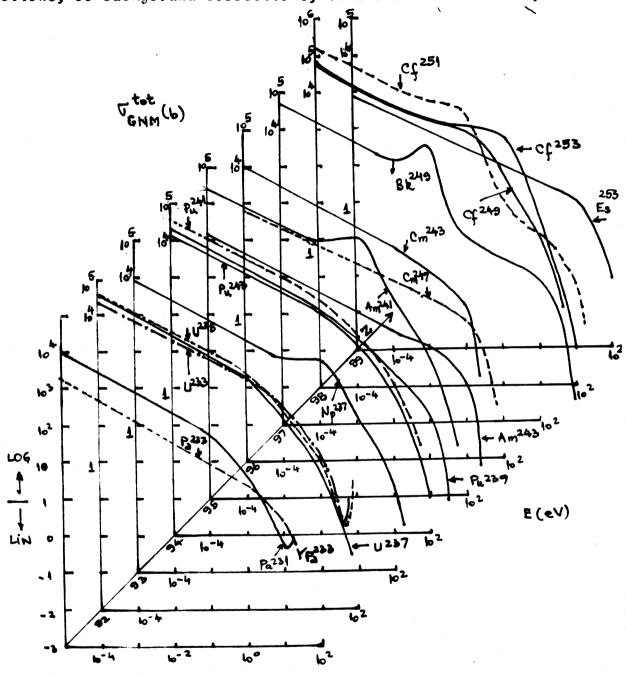


Figure 1
The contributions of the truncated level for total cross sections of the transactinides with odd mass numbers

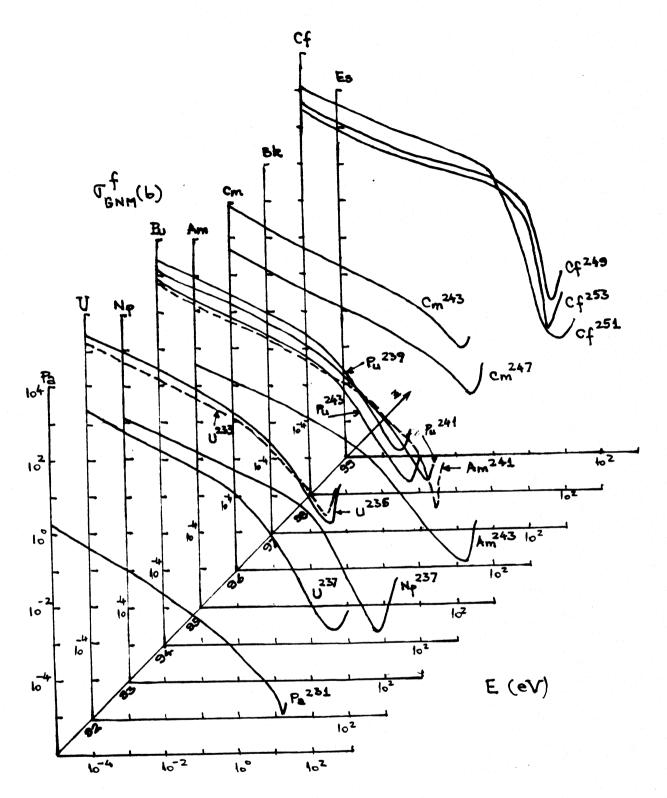


Figure 2

The contributions of the truncated level for fission cross sections of the transactinides with odd mass numbers

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Hydrogeological investigations using the neutron activation analysis in tracers methods

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The neutron activation analysis associated with the chemical preconcentration of the elements by coprecipitation reactions followed by the filtration on the nuclear membranes, proves to be a highly sensitive analysis method. This method has been improved and intensely applied by the authors in various hydrogeological investigations like: study of the dynamics of underground and surface waters in karstic zones, investigation on the infiltrations in mines, investigation on the exfiltrations from accumulation reservoirs, etc.

In, I, Dy and La have been used as activable tracers in the following compounds: In-EDTA, KI, Dy-EDTA, La-EDTA and presently work is beeing carried on the develop the use of Br in the NH_Br compound.

The main nuclear and analytical parameters of the activable tracers used, are given in the following table:

Ele- ment	Isotope used and relative abundance (percent)	Thermal neutrons activa- tion cross section (b)	Measured gamma ray (keV) and intensity	Chemical stabili- ty coeff. (lg K _s)	Detection limit ob- tained (g/g)
In	115In(95,67)	155	417,0(0,30)	24,9	0,9.10-12
I	127 I (100)	6,2	442,9(0,179)		1.10-10
Dy	164 Dy (28, 10)	800	94,6(0,04)	18,3	2.10-13
La	139 La (99,91)	8,8	1596,2(0,96)	15.5	1.10-10
Br	79Br(50,56)	8,5	617,0(0,072)		1.10-9

The neutron activation has been carried out by means of an air rabbit at the nuclear reactor in a neutron flux of 10^{12} n/s.cm² and the spectroscopical determinations, by using a Ge(Li) and a GeHp and 4096 channels analyzer.

In a period of 5 years, 45 labellings have been performed for underground and surface water flows with a debit of 0,1-30 cubic meters per second and transit times of up to 200 days. Two ac-

cumulation reservoirs, one of 0.3 million and one of 6 million cubic meters, have been investigated.

The method of activable tracers proved to be complementary to chemical and radioactive tracers methods, but in some cases superior to them from the point of view of the environmental protection and of the possibility to label some waters of high volume and long transit time.

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Analysis of high purity semiconductor silicon by neutron activation

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Analysis of the impurities contained in high purity semiconductor materials is an important problem since they might drastically change their mechanical and electrical properties.

Quantitative determination of trace elements in semiconductor silicon is obtained at VVR-S reactor, Bucharest using neutron activation method. Many samples of semiconductor silicon slides of various types (p or n) and different sorts (Romanian or foreigner) are analysed. Before the irradiation in a 1.2×10¹³ n/cm².s flux the silicon slides were cleaned and washed. As, Au, Co, Cr, Fe, Ga, Hf, Mo, Na, Ni, Sb, Sc elements were found out.

The results expressed in atoms/cm² reveal a surface contamination of the silicon slides during cutting, polishing, washing and handling before and after their irradiation. For this surface containination values of $(10^{10} \cdot 10^{15})$ atoms/cm² are obtained.

A study of the variation of some elements content with succesive etchings of the samples after irradiation has also been made. After the first etching a high decreasing of the element contents can be observed. An etching of 5 minutes in a 5:3:3 mixture of acids (HNO₃, HF, CH₃COOH) reveals the volume contamination of silicon semiconductor slides. The values of (10¹¹ *10¹⁴) atoms/cm³ for this volume contamination are obtained.

Rare earth analysis of some geological samples by neutron activation

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Determination of rare-earths contents in rocks and minerals is of great importance in geochemical and crystallochemical studies to appreciate the origin and evolution of various types of rocks. It can also be of practical importance of mineralogical exploitations point of view.

Many geological samples from the eastern Carpathians have been analysed or these analyses are in progress. The samples are irradiated in VVR-S reactor in a 2×10^{12} n/cm 2 ·s flux. GSP-1 and Soil-7 are used as standard materials. The measurements have been carried out by using a Ge(Li) detector (2.2 keV resolution).

Concentration of subppm or tens of ppm for Ce, Dy, Eu, Gd, La, Lu, Nd, Sm, Tb, Yb rare-earth have been obtained.

In addition, contents of Au, Ba, Ca, Co, Cr, Fe, Hf, Mn, Na, Rb, Sc, Sr, Ta, Th and U have been determined. The concentration of these elements are widely spread between subppm and percents.

Neutron activation analysis of some marine algae and phanerogams along the Romanian Black Sea coast

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Due to the intense concentration ability of chemical elements by the marine macrophytes from the sea-water, the study of their elemental composition is of interest for algological and environmental studies. The knowledge of the chemical composition of marine flora is also an useful and inexpensive method for metal contamination evaluation. Thus, some species can be used as environmental indicators for different chemical elements. The concentration of 23 elements contained in 11 species of marine macrophytes algae and marine phanerograms collected along the Romanian Black Sea coast has been determined. Enteromorpha linza, Ulva lactuca, Bryopsis plumosa, Cladophora vagabunda, Cladophora sericea (Clorophytes); Cystoseira barbata, Scytosiphon lomentaria (Phaeophytes), Ceramium elegans, Porphyra leucosticta (Rhodophytes), Zostera nana, Zostera marina (Phanerogams) have been analysed.

The washed, dried $(105^{\circ}C)$ and ashed $(480^{\circ}C)$ samples have been irradiated in a thermal flux of 1.1×10^{11} n/cm²·s of VVR-S reactor. The measurements have been carried out by using a 65 cm³ Ge(Li) detector with 2.2 keV resolution.

The concentration of major constituents Na, K, Ca, Mg, Fe, Al, Sr (%) and Mn, Zn, Ba (hundreds ppm) were determined. For Ce, Co, Cr, Cs, Eu, Hf, La, Lu, Rb, Sc, Sm, Th, Yb elements the concentration of 10 ppb + 10 ppm has been obtained. V was found only in two samples and Ti in one sample. The variation of 3+10 times of the elemental concentration for different species analysed was obtained.

The seasonal variation of concentration has also observed.