Institute of Isotopes, Hungarian Academy of Sciences
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The **fundamental tasks** of the Institute are (by the Institute's statutes):
to conduct internationally recognized scientific research in the fields of

- radiation effects (induced by neutrons, γ-rays and electrons),
- isotope and nuclear chemistry,
- radiation chemistry
- radiation protection and safety,
- reaction kinetics, and
- heterogeneous catalysis.

Furthermore, the Institute should provide advisory and service activities in relation to the promotion and accomplishment of the implementation of the legal control of national utilization of the atomic energy.
In addition, the Institute

- takes part in higher education (gradual and post-gradual level),
- conducts advisory activity, and
- may perform other occasional R & D services.

The total number of the personnel is 92, the half of which is research fellow.
The year's total budget is c.a. 2 M € (covered in c.a. 2/3rd part by direct financing provided by the Hungarian Academy of Sciences).

The research work is pursued in five departments:
1) Radiation Chemistry,
2) Nuclear Research,
3) Radiation Safety,
4) Surface Chemistry and Catalysts
5) Catalysis and Tracer Studies

A short description of their activities:

1. Department of Radiation Chemistry

Research Areas

The main research areas are:

- Radiation and photochemistry of hydrocarbons.
- Degradation of environmental pollutants e.g. chlorinated aromatic hydrocarbons, reactive textile dyes.
- Polymer radiation chemistry and mechanism of radiation induced polymerization.
- Radiation methods for synthesis of polymers for biomedical applications.

Short term research aims

- Kinetics of radiation induced polymerization of acrylamide, acrylate and methacrylate type compounds in aqueous solutions and in cyclohexane.
- Radiation induced transformation in cellulose using textile, wood samples and polysaccharides.
- Synthesis of hydrogels for biochemical applications, e.g. for slow release of drugs, wound dressing.
- Production of functional polymer micro- and nanoparticles for purification, separation, diagnostic and drug delivery purposes.
- Radiation induced degradation of phenolic and chlorinated aromatic hydrocarbons and azo dyes in aqueous solutions.

Methodology and Instrumentation

- The department makes use of and automated pulse radiolysis facility with a 4 MeV LINAC and a 300 keV RADAN accelerators, and a semi-industrial ^{60}Co irradiator facility as well.
- A conventional analytical laboratory (UV-VIS spectrophotometers; FTIR spectrophotometer with transmittance, ATR, DR detectors; GC and HPLC with diode array detector).
- A plasma surface treatment equipment.
- A JEOL JSM-5600 LV type scanning electron microscope with EDS and XRF detectors.

2. Department of Nuclear Research

Research areas

The main research areas are:

- Prompt Gamma Activation Analysis (PGAA) and its applications
- Nuclear spectroscopy with neutron-induced reactions
- Nuclear data measurements and evaluation
- Gamma-ray spectrometry and metrology
- In-beam Mössbauer spectroscopy

Short term research aims

- Re-measurement of spectroscopic atlas of neutron-induced prompt gamma rays of 80 elements for prompt gamma analysis with higher neutron flux.
- Application of PGAA for determining the elemental compositions and revealing the impurities in various catalytic materials and to investigate the dynamics of catalytic reactions.
- Non-destructive determination of elemental compositions of various archaeological objects by PGAA to determine provenance, trading routes of the objects.
- Run EU user program in the framework of “The Integrated Infrastructure Initiative for Neutron Scattering and Muon Spectroscopy” project of EU FP6.
- Study elemental compositions, especially boron content of volcanic rock and geological samples.
- Development of in situ PGAA.

- Development of a PGAA based method for the measurement of isotope ratios in fissile materials and presence of fissile material in container or in presence of masking radioactive sources.
- Thermal neutron-capture cross section measurements of LLFF nuclear wastes, minor actinides, fissile materials and target materials of accelerator driven systems for transmutation in co-operation with EU JRC IRMM.
- Data improvement of energy and efficiency calibration standards for high energy gamma ray detector calibrations.
- Installation and test experiments with the new in-beam Mössbauer spectrometer.
- Development of neutron optics and of 3D imaging with slow neutrons.

Methodology and Instrumentation

- Experimental activities are based on the 10-MW Budapest Research Reactor, equipped with a cold neutron guide system and a liquid hydrogen cold neutron source. The Institute is a cofounder of the Budapest Neutron Centre, an international neutron user community.
- Two experimental stations on divided neutron guide for PGAA and other research, utilising neutron-induced gamma-ray spectroscopy as a tool. The bent guide, robust radiation shielding and evacuated sample chamber provide excellent background conditions, hence a high signal-to-noise ratio.
- Gamma-ray spectrometer consisting of 21% HPGe detector guarded by an eight-segment BGO annulus and BGO back-catcher crystals. It can be operated either in Compton-suppression or annihilation-pair modes, to collect 16–64 thousand-channel spectra.
- Additional HPGe semiconductor detectors, associated electronics and a multiplexer data acquisition system for gamma-gamma coincidence spectroscopy.
- Two BaF_2 scintillators for fast coincidence-timing measurement in nuclear spectroscopy.
- A liquid helium cooled in-beam Mössbauer cryostat built on a converging super mirror neutron guide. It is equipped with all nuclear instruments necessary for performing high quality Mössbauer spectroscopy.

3. Department of Radiation Safety

This Department consists of four Sections, namely: Nuclear, Dosimetry, Radioactive Material Registry Sections and ICP-MS Mass Spectrometry Laboratory

3.1 Nuclear section

Areas of main activities

The main research fields and related activities are:

- Nuclear safeguards
- Combating illicit trafficking of nuclear materials

Recent results and current research

Determining the nuclear-material content of a mixture of damaged spent fuel within sealed containers at Paks NPP. Due to an incident at the Paks NPP in 2003, 30 fuel assemblies kept in a “cleaning tank” became damaged. Many fuel rods broke and different types of pellets fell out from the rods and got mixed with each other and with broken construction elements (e.g. cladding). The damaged fuel will be repackaged into about 100 closed canisters and kept in the spent-fuel pond until further action. Declaring the nuclear-material content of the individual closed canisters is a difficult task, because the canisters will contain a mixture of spent-fuel pieces of different initial enrichment, burn-up and irradiation history distributed in an irregular geometry. A passive nondestructive method has been developed at the Department of Radiation Safety which will be used for determining the nuclear-material content of the closed canisters at the Paks NPP. The method is based on the combination of gamma-spectrometric and neutron counting measurements. These measurements will provide the inventory to be declared. Although various equipment using the combination of gamma spectrometry and neutron counting for the verification of regular spent-fuel assemblies already exists (e.g. the SMOPY and EFDET devices), presently they are not capable of determining the mass of nuclear material of such an irregular structure, as the structure of the damaged fuel at the Paks NPP. Therefore specific equipment will be built and a novel methodology will have to be applied. The measurements to be carried out could serve as a precedent for inventory-taking of nuclear material in a mixture of different types of nuclear and non-nuclear material distributed in an irregular, unknown geometry.

Quantitative assay of plutonium-beryllium sources. Pu-Be sources represent a nuclear safety and safeguards issue since they contain nuclear (fissile) material. A large number of such sources – mostly out of use – are stored in Hungary (and this is the case in several neighbouring countries as well). The Pu content of these sources is to be accounted for, regularly reported to and inspected by IAEA, and from 2004 on, also by EURATOM safeguards. Since the late Soviet supplier did not declare the Pu-content of the sources, the values for the Pu content presently on record are based on rough overestimates assuming that the sources contain pure ^{239}Pu. Recently, the isotopic composition of some of the sources was determined by gamma-spectrometry and more precise estimates for their Pu content were given. These estimates, however, still included the high uncertainty associated with the specific neutron yields needed for the calculation of the Pu content. In parallel with the gamma-spectrometric measurement of the isotopic composition, a high-resolution gamma-spectrometric method which does not require the knowledge of the specific neutron yields was developed for the experimental determination of the Pu content of these sources. Furthermore, combining the results of the gamma-spectrometric measurement with the measurement of the total neutron counts the specific neutron yields of the sources can also be determined, thus providing additional information about the sources. The gamma-spectrometric method made it possible to prepare a set of calibrating sources which were used in developing an independent neutron coincidence counting method for the determination of the Pu content of the sources. A neutron coincidence counter was built and a correlation was obtained between the Pu-content and the ratio of the number of real coincidences to the total number of counts. This method provides another alternative to determine the Pu content and also to derive additional information about the sources. This research was accepted by the IAEA as part of the Hungarian support programme to the IAEA. Recently, in collaboration with JRC Ispra, the results of the Pu mass measurements of several sources were confirmed by calorimetric measurements.

Assistance to IAEA inspectors during inspections in the nuclear power plant in Paks, Hungary. A portable spent fuel attribute tester (PSFAT) for the verification of WWER type nuclear fuel assemblies was developed by the safeguards group of the Department and it is now in routine use at the Paks power plant. The attribute tester is based on medium resolution gamma spectrometry using a CdZnTe detector. The equipment is primarily designed for distinguishing between low-burnup spent-fuel assemblies, absorbers and Co-containers stored in the spent fuel pond (even in the neighbourhood of other strong gamma-emitting assemblies), without the need to move the assemblies. In this way the attribute tester can help the safeguards inspectors in the situations when Cherenkov viewing devices are not usable. The group has also experience in using high-resolution gamma spectrometry and neutron counting techniques for examining reactor fuel rods.

Combating illicit trafficking of nuclear materials. Due to its geographical location, Hungary is a transit country on the route of illicit trafficking of nuclear materials. During the 1990s there were several events in Hungary involving the seizure of such materials. According to Hungarian legislation, the Institute of Isotopes is responsible for *on-the-spot and laboratory analysis of nuclear material of unknown origin*. In order to decrease the threat of “nuclear terrorism” it is of paramount importance to find out as much as possible about the nuclear materials coming in this way into Hungary, as well as about their possible origin. Several methods using high resolution gamma-spectrometry for the non-destructive assay of unknown nuclear materials were developed and are being developed in the Department of Radiation Safety. For example, a unique method for determining *the matrix of uranium samples* of unknown origin was developed. It relies on the gamma-spectrometric measurement of the total uranium content of the samples.

Development of a novel gamma-spectrometric method for uranium age dating. The age of uranium samples is an important piece of information relevant both in combating illicit trafficking and in nuclear safeguards. It can help in determining the origin of seized or found nuclear materials, and, on the other hand, knowing the date of production of the material can help the safeguards expert to decide whether a highly-enriched uranium sample originates from excess weapons-usable materials or it is freshly produced. The method for age dating relies on measuring the daughter/parent activity ratio $^{214}Bi/^{234}U$ by low-background, high-resolution gamma-spectrometry. The initial methodology was derived during a "Round Robin" exercise, in which the properties of a HEU material relevant to nuclear forensics were assessed by several laboratories. The uranium-age obtained by this gamma-spectrometric method was in agreement with the results reported by other participating laboratories, which used mass-spectrometry. The original method relied on using an efficiency calibrated geometry, which was easily achieved for thin samples in powder form. Since then, using a uranium sample of medium enrichment, the method was extended to determining the age of uranium material of any physical form and geometrical shape. This is made possible by using the peaks of ^{238}U for relative efficiency calibration, provided that they can be evaluated from the spectrum. In the present measurement setup (with a 150 cm^3 coaxial HPGe detector) the lower bound for age determination of 90% enriched HEU was estimated to be around 5 years. If the presently best available well-type HPGe detector could be used, the sensitivity of the method could be improved, so that the lower bound for the applicability of the method would be approximately 1 year for 90% enriched uranium and 20 years for natural uranium.

Instrumentation

- A range of high-purity germanium detectors is available (three planar detectors with diameters of 10, 36.7 and 50.5 mm and two coaxial detectors with volume of 46 and 150 cm^3) for high-resolution gamma spectroscopy.
- A low-background iron chamber is used for the gamma-spectrometric measurements of low activities (e.g. for uranium age determination).
- Medium resolution gamma-spectroscopy is performed using a small volume (60 mm^3) and a large volume (500 mm^3) CdZnTe detector.
- For neutron coincidence counting a dedicated device was built, consisting of 9 pieces of CHM-18 type proportional counter tubes filled with ^3He, placed in polyamide moderator. The device uses home-made electronics and a JSR-11 type shift register (lent to the Institute by the IAEA).

3.2 Dosimetry Section

Areas of main activities

The main research fields and related activities are:

- Chemical dosimetry
- Solid state dosimetry
- Environmental and personal dosimetry
- Accidental and reactor dosimetry
- Radiation processing dosimetry
- Dosimetry control at industrial high activity gamma and at high energy electron irradiation facilities
- Radiation protection services

Short term research aims

- Development of dosimeter systems based on measurement of optically stimulated luminescence (OSL), conductivity and/or light absorption/reflection for environmental, radiation therapy and radiation processing purposes; investigation of the conditions of usability.
- Development of passive thermoluminescent (TL) dosimetry systems with high sensitivity and/or LET selectivity to measure dose of mixed low and high LET radiation fields i.e. to monitor aircraft and spacecraft personnel and to measure dose in the stratosphere.
- Development of use of TL (Al_2O_3:Y, Mg; Hungarian development) and OSL (Sunna film) dosimetry systems to measure dose at room- and high (~100°C) temperature at nuclear power plants e.g. in difficult–to-access areas ("hermetic zones").
- Investigation of the thermoluminescence-, radioluminescence- and optically stimulated luminescence properties of EU doped materials and materials (mainly quartz and feldspar) used in retrospective and environmental dosimetry.

Methodology and Instrumentation

The experimental methods involve the followings:

- Different types of optical spectroscopies (optical absorption, photoluminescence, optically stimulated luminescence), various thermally stimulated methods (thermoluminescence [TL], phototransfer TL, thermally stimulated exoelectron emission, thermal bleaching) and radioluminescence (RL). Emphasis is placed on the simultaneous measurements of the above characteristics (e.g. thermoluminescence vs. thermal bleaching of optical absorption; radioluminescence vs. thermoluminescence).
- A computer controlled vacuum-cryostat is also available for both low and high temperature measurements.
- The environmental and personal dosimetry research and use applies mainly thermoluminescent evaluation on LiF, LiF:Mg,Ti, Al_2O_3:C, $CaSO_4$:Tm, Li_2B_4O7, $Li_2B_4O_7$:Cu and $CaSO_4$:Dy dosimeters.
- Oscillometry, conductometry and reflectometry partly with locally developed routine readout instruments.
- The dosimeter systems applied in radiation processing involve the reference standard Fricke solution (UV absorption spectrophotometry), the reference standard ethanol monochlorobenzene solution (non-destructive oscillometric evaluation), water, graphite and polystyrene calorimeters, the Perspex (PMMA) dosimeters (visible spectrophotometry), radiochromic films (FWT-60, B3, GafChromic – all visible spectrophotometry) and the LiF/PE Sunna dosimeter film (OSL analysis).
- A wide variety of irradiation facilities including research and industrial scale gamma (e.g. 3.7 PBq ^{60}Co source) and electron (e.g. 4 MeV linear electron accelerator) irradiation facilities and neutron sources are used for these investigations.

3.3 Radioactive Material Registry Section:

Areas of main activities

- Center registry of radioactive materials in Hungary with computer system. The database contains all radioactive materials produced in or imported to Hungary since the beginning (1954) of the domestic use of artificial radioactive products.
- Software development for the central registry.

3.4 ICP-MS Mass Spectrometry Laboratory

High resolution mass spectrometry has recently been implemented at the Institute. The primary objective of the project was to complement traditional NDA techniques by DA capabilities used for the identification and measurement of nuclear materials.

Supported by PHARE funding an ELEMENT 2 ICP-MS was installed in clean laboratory environment.

Main R&D areas

- Analysis of nuclear material of unknown origin (illicit trafficking). Determination of isotopics of uranium and other actinides, analysis of trace elements
- Assessment of nuclear material collected on filters and swipes, or contained in other safeguards related samples
- Analysis of long-lived radionuclides in environmental samples
- Study of the laser ablation technique in the above and other fields

Instrumentation

- ELEMENT 2 ICP-MS (Finnigan)
- Various sample introduction systems (e.g. ARIDUS, micro-Sampler, Stable sample Introduction System, Microconcentric PFA Nebulizer incl. ASX-100 Autosampler)
- Laser Ablation System (UP 213, New-Wave Research)
- Various sample preparation equipment (e.g. MARS-5 Microwave Sample Digestion, CEM)

Note
In other laboratories of the Institute of Isotopes and other institutes within the research campus there are a number of related analytical techniques, such as XRF, SEM, NAA, PGAA, PIXE, alpha-spectrometry.

4. Department of Surface Chemistry and Catalysis

Research areas
Molecular level investigation on the development of highly dispersed, nanoscale mono- and multimetallic particles deposited over various supports of different structures (sol/gel, zeolites, oxides, single crystals, etc). Studies on the modification of interfacial structure and structure/reactivity relationship using highly sophisticated physical (XPS, UPS, STM, FTIR, etc.) and chemical (adsorption, isotope exchange and catalytic reactions) methods.

The main research areas are:
- Studies on surface of highly dispersed metal particles: model catalysis
- Activation of C_1 molecules: structure and reactivity relationship of promoted mono- and bimetallic catalysts
- Selective hydrogenation
- Environmental Chemistry

Short term research aims:
- Model catalysis: Modification of the interfacial structure between gold nanoparticles and oxides (e.g. Fe_2O_3, TiO_2, MoO_3) and the effect of second metal. Study of the respective structure and catalytic activity relationship of well characterized gold and modified gold nanoparticles. Study the role of nanosize Pd particles in Pd-Ag/pumice catalysts in CO oxidation.
- Homologation of CH_4 in the presence and absence on N_2O as well as CO hydrogenation over bimetallic catalysts. XPS and TPR characterization of H-ZSM-5 supported Ga and transition metal containing bimetallic catalysts utilized for production of high molecular weight hydrocarbons.
- Study the mechanism of DeNOx reaction on noble metal catalysts using transient kinetics and FTIR method.

Methodology and instrumentation

- Flow-through and CSTR reactors for medium pressure (max. 40 bar), atmospheric and sub-atmospheric pressure operation.
- Catalyst characterization in flow system: chemisorption, TPD, TPR in the SORBSTAR facility (developed and constructed by the Department),
- Catalyst characterization by surface science techniques: Kratos ES-300 equipped with XPS, AES and UPS sources, with preparation chamber: Kratos XSAM pci XPS with atmospheric reaction chamber; STM combined with LEED, AES, TPD.
- Midac M010H FTIR spectrometer with catalytic cell.
- QMS-s for transient labeling, on-line analysis and isotope exchange.
- DuPont 21-490 GC-MS combined with radio gas chromatographic attachment
- Hiden QMS for isotope exchange.
- Packard gas chromatographs.
- Perkin-Elmer DSC-4.
- Sartorius vacuum microbalance for adsorption measurements.

5) Department of Catalysis and Tracer Studies

Research Areas

The activities exerted on the Department fall into two main directions: one is the study of heterogeneous supported catalysts; the other is the specific application of radiotracers. Nevertheless, these two branches of research are connected to each other, as ^{35}S is used for identifying various types of sulfur present on alumina/molibdena supported catalysts, and Mössbauer spectroscopy is applied for characterizing catalysts containing iron or tin modifiers.

The approach applied for studying supported bimetallic catalysts is strongly based on the preliminary knowledge on the correlation existing between the catalyst structure and actual reaction pathways. Therefore, the basic tool used for characterizing the catalysts is the selectivity of desired catalytic reactions. This kind of characterization is completed with other surface specific (AES, work function, XPS) and conventional (TPR, H_2/O_2 titration, surface area) methods.

The main research areas are:

- Study of supported bimetallic catalysts in the conversions of hydrocarbons. Modification of the active Pt, Rh and Pd particles by adding inactive elements, such as Ge, Sn, or active ones such as Pd, Rh or Pd by controlled surface reactions (SCR), either via organometallic compounds or by underpotential deposition. The occupation and selective blocking of different sites and the interacting effect of the two active phases is being investigated.
- The structure, catalytic effect (influence on the activity and selectivity pattern) of carbonaceous deposits formed during refinery model reactions (skeletal rearrangement of C_6 hydrocarbons) is studied and correlated with the experimental conditions of their formations.
- Study the selective oxidation of CO (PROX) in presence of hydrogen for fuel cell application, using different CeO_2 and CeO_2-ZrO_2 mixed oxide supported metal catalysts.
- Identification of various sulfur species (removable, exchangeable and strongly bonded) and study of their interconversion on sulfided VIIIB metal containing supported catalysts applying on-line beta detection of ^{35}S tracer.

- Study of surface species formed on platinum upon chemisorption of low molecular weight hydrocarbons applying Auger spectroscopy (AES) and work-function measurement at UHV conditions.
- Identification of various oxidation and coordination states and study of their interconversions in iron or tin containing catalysts by in situ Mössbauer spectroscopy.
- Study of migration of long half-life radioisotopes typical of high-level nuclear waste in geological samples (borecores).

Short term research aims

- Bimetallic catalysts: study of the modifying effect of Ge and Sn on supported Pt and Rh catalysts, study the interaction of Pt-Pd, Pt-Rh using hydrocarbon transformation as test reactions; introduction of new possible test reactions, other than hydrocarbon transformations, such as selective hydrogenation of acrolein.
- Comparative study of Pt/CeO_2 and Pd/CeO_2 catalysts in the PROX reaction.
- Study of surface species formed on Pt from chemisorption of ethanol and furan by AES and work-function measurements.
- Study of the oxidation and coordination states of iron in micro- and mesoporous ferrisilicates.
- Break-through measurements for determination of apparent diffusion constant of anionic species ($^{99}TcO_4^-$, $^{125}I^-$ and $H^{14}CO_3^-$) in argillaceous borecore samples.

Methodology and instrumentation

- Different flow-through and closed-loop catalytic reactors equipped by gas-chromatographs
- Catalyst testing by chemisorption: TPR, TPD, surface area, O_2-H_2 titrations and catalytic reactions performed in the same integrated system,
- Three CHROMPACK 9001 and two Packard 427 and other gas chromatographs with capillary and regular columns with TCD & FID.
- On line flow-through scintillation cells for detection of beta radiation of ^{35}S and ^{14}C tracers.
- UHV system (work function, flash desorption, MS detection of products, Auger option)
- Mössbauer spectroscopy (in situ cell operating in 77–700 K range for measuring and/or sample treating)
- Mettler TA-1HT computer controlled thermobalance (TG and DTA up to 1200 K)
- gamma spectrometry with HPGe detector (OXFORD CNVDS30-25185)
- high pressure catalytic microreactor (Parr, up to 20 MPa)

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Further updated information and more details on the activities of the Departments are available on the web site of the Institute (http://www.iki.kfki.hu; in particular, the list of publications authored/coauthored by the Institute's personnel, or the list of personnel and connections to the staff are shown as well.)