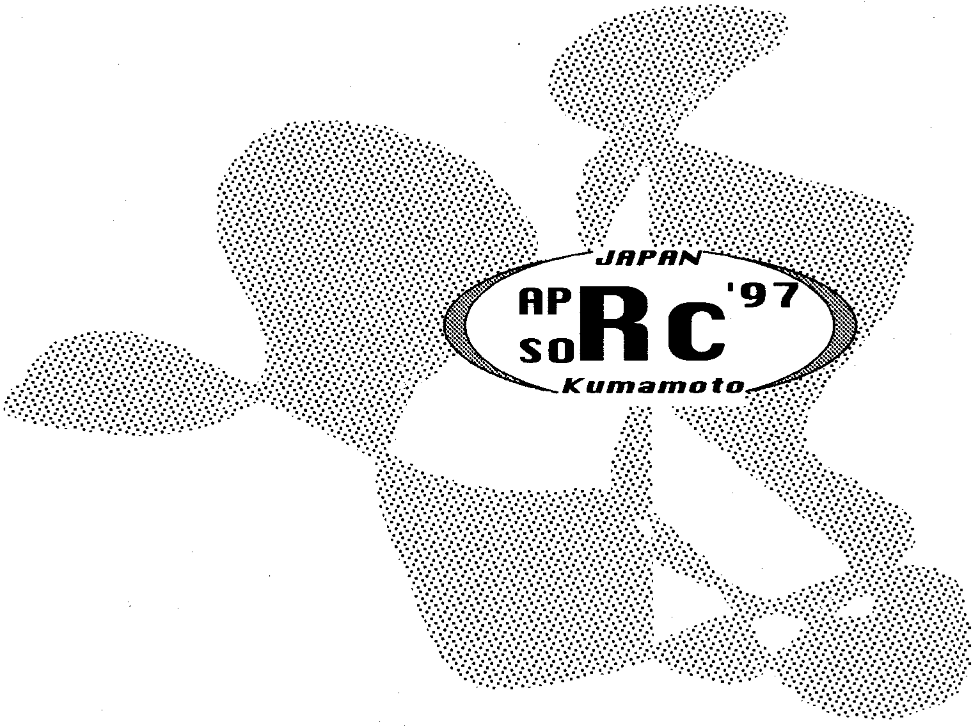
APSORC'97 ASIA-PACIFIC SYMPOSIUM ON RADIOCHEMISTRY-KUMAMOTO, JAPAN

APSORC '97 LIST OF ABSTRACTS

PLENARY LECTURES



- KEYNOTE THEME -

**EVER ONWARD TOWARDS THE FRONTIERS OF
RADIOCHEMISTRY IN THE
SECOND CENTURY OF RADIOACTIVITY DISCOVERY**

PL1
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NEUTRON ACTIVATION ANALYSIS CHALLENGES:
PROBLEMS AND APPLICATIONS IN BIOMEDICAL AND OTHER AREAS.

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Summary: Developments in neutron activation analysis in its various modes for trace element determination are discussed with reference to reactor and other sources, competing methods, tomography and detection systems in biomedical research.

Key Words: Neutron activation analysis,trace elements,tomography, biomedical.

Neutron activation analysis (NAA) is a well established technique for the determination of elemental concentrations at trace and ultratrace levels in a variety of matrices. It has been applied to diverse problems in scientific and technological areas and has proven extremely valuable in the certification of reference materials and quality assurance procedures.

The compound nucleus formed on capture of a neutron by the target nucleus, de-excites by the prompt emission of characteristic gamma-rays which serve to identify the element (NCPGA). The radionuclides which may result after neutron irradiation of a sample also emit characteristic gamma-rays following β^- decay and their spectrum is recorded for calculation of elemental concentrations. The half-lives of the radionuclides are broadly divided into short-lived and long-lived with the latter providing the possibility of radiochemical separation in order to enhance detection limits by removal of unwanted matrix effects and interferences (RNAA). Developments in rapid radiochemical separation processes for short-lived nuclides have taken place but the emphasis in the main has been on instrumental methods (INAA) for improved detection. INAA encompasses cyclic and pseudocyclic neutron activation analysis (CINAA and PCINAA) with or without Compton suppression detection, combined with rapid transport systems which reduce transfer times (<1s) between irradiation and counting positions thus minimising the loss of detectable events through decay. A parallel development in the measurement of high activity sources is the design of electronic instrumentation for 'loss-free' counting spectroscopy systems.

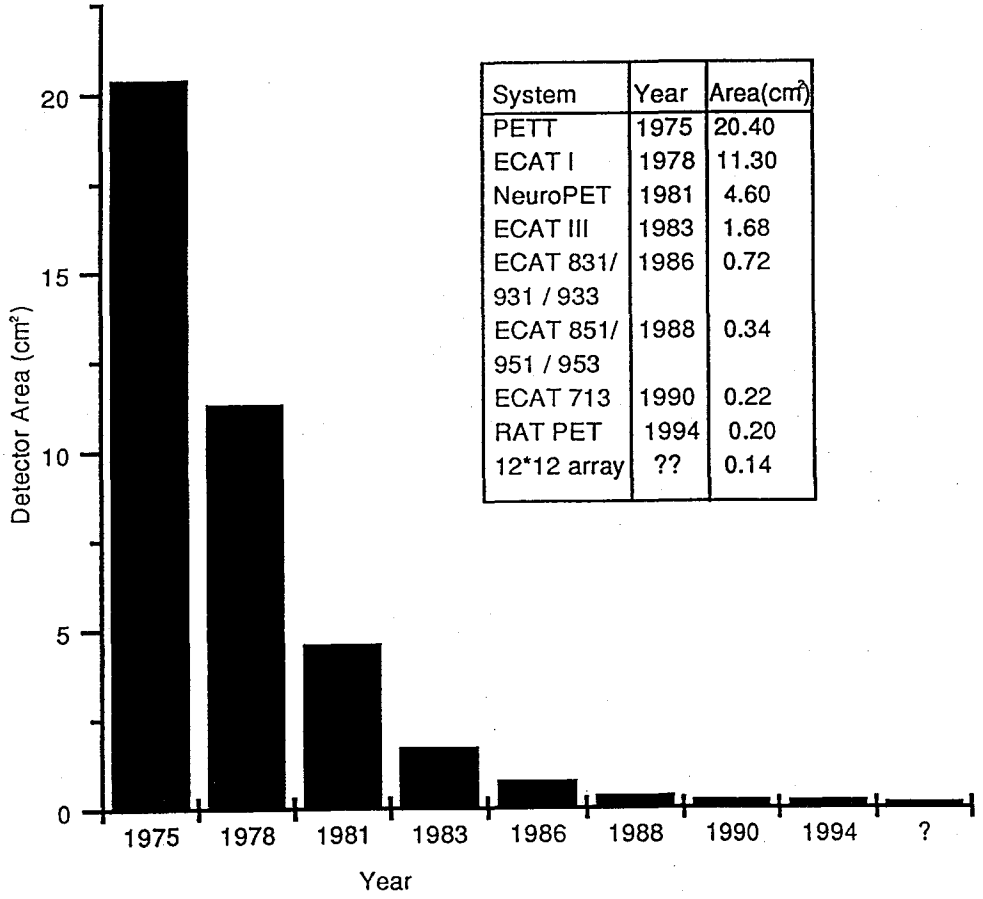
Other multielemental analytical methods have come to the forefront in recent years, notably inductively coupled plasma mass spectrometry (ICPMS) and atomic emission spectrometry (ICPAES). Rapid turnaround and very good sensitivities for a large number of elements and matrices have encouraged the considearable growth in the use of these analytical instruments worldwide, in a relatively short period of time. Freedom from radiation hazards is considered an additional attraction. However, radiation-based analytical methods, are also being increasingly employed. Ion beam analysis facilities, now specifically designed for the purpose and not as of old converted from accelerators used in nuclear physics research, are being installed largely spurred by research in the microelectronics and plastics industries. Proton induced x-ray emission (PIXE) analysis, Rutherford Backscattering (RBS) and other charged particle reactions are powerful tools which with microprobe scanning facilities and stable ion beams provide not only the means to determine elemental distribution in a sample but also to analyse large batches of samples in sequence. X-ray fluorescence (XRF) systems are becoming available to a larger radioanalytical community due to relatively less expensive equipment, allowing work to be carried out, for example in environmental and agricultural studies, at centres where these have not been hitherto possible. There is a need for these sometimes competing analytical techniques to be evaluated and compared, common problems and challenges be identified, complimentarity and uniqeness discussed and established. One such forum for discussion was at the recent (November 1996) IAEA Symposium on 'Harmonisation of health related environmental measurements using nuclear and isotopic techniques', in Hyderabad, India.

The cost effectiveness and ageing of research nuclear reactors have been under financial scrutiny in the last decade and in particular in Europe and North America. This has led to rationalisation in the number of reactors where, for example, in the UK two out of three Universities research reactors have been closed down and decommissioned. Some centres though have risen to the challenge and expanded their facilities, notably developing cold neutron guide beams for neutron capture prompt gamma-ray analysis (NCPGA) eg. in Hungary, USA and Japan, and systems for the irradiation of really large objects or samples (the Netherlands). There has also been a re-examination of the usefulness of thermal and epithermal NAA with appropriately tailored neutron spectra. However, new research reactors have also been installed such as the ones in South Korea, Ghana and Syria. In the last two countries these are the new generation of Chinese turn-key reactors almost exclusively dedicated to activation analysis.

NAA is not the exclusive province of reactor based systems as both isotopic neutron sources and neutron generators have made a significant contribution to well defined fields. These include 'in vivo' NAA, nuclear safeguards, detection of explosives and field analysis of minerals and raw materials. The lower neutron flux in these applications is generally compensated for by the larger samples, the versatility of irradiation and counting geometry, transportability and in 'in-vivo' NAA optimisation within the constraints of absorbed dose. The development of small neutron generators, for incorporation into neutron interrogation systems provides exciting possibilities for applications in wider areas. A re-evaluation of employment of isotopic sources is necessary, particularly in biomedical applications where the impetus is provided by research in boron neutron capture therapy. This has been recognised with the establishment of the Californium User Facility, ORNL, USA based on compact Cf-252 fission neutron sources.

INAA in its prompt and delayed modes, determines the concentration of elements in a whole sample, nondestructively. If the sample is rare or unique this has considerable advantages, not least because another analytical technique can be used to obtain additional information. However, it is also possible to combine the sensitivity of NAA with the principles of computerised tomography and obtain the distribution of elements in the sample, constructing a 3-D map. The technique is termed neutron induced gamma-ray emission tomography (NIGET). With a single collimated high resolution Ge detector acquisition of tomographic data is time consuming because of the low detection efficiency. The use of multiple arrays of BGO detectors, developed for positron emission tomography (PET) based on the coincident detection of annihilation photons, is being considered, especially as the surface area of each detector has decreased significantly over the last 20 years, Figure 1, allowing good spatial resolution. But the energy resolution of the detectors is poor making discrimination difficult between neighbouring gamma-ray lines. Cascade gamma-ray coincident tomography may obviate some of the problems and will be illustrated. Additionally the possibility of determining the chemical binding of a radionuclide emitting gamma-rays in cascade will be discussed.

Biomedical areas of research in which NAA has a role to play will be presented and include Alzheimers disease, diabetes mellitus, scar formation, gastrointestinal tract disorders, brain-gut interactions and mineral supplementation.



System	Year	Area(cm²)
PETT	1975	20.40
ECAT I	1978	11.30
NeuroPET	1981	4.60
ECAT III	1983	1.68
ECAT 831/ 931 / 933	1986	0.72
ECAT 851/ 951 / 953	1988	0.34
ECAT 713	1990	0.22
RAT PET	1994	0.20
12*12 array	??	0.14

FIGURE 1 Area of detector face in successive generations of Positron Emission Tomography (PET) systems.

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PL2
s.61

THE PARITY METHOD FOR ACTIVITY MEASUREMENTS

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Summary: A new method for absolute activity measurements is proposed. Instead of measuring coincidences one determines, by modulo 2 counting, the probability that over a given short time interval the sum of the registered beta and gamma events is an odd number.

Key words: Radioactivity, Coincidence method, Modulo 2 counting.

We briefly introduce the parity method, a novel approach to absolute activity measurements, and present its current state of development. After describing the simple idea underlying the method, the complications arising from the need to take account of dead times are discussed. This calls for some mathematical elaboration but does not affect the principle. One may wonder why a new method should be developed. The activity of a source is traditionally measured by the coincidence method, provided that the decay proceeds by the emission of two distinct radiations, normally a beta particle and a gamma ray. This method is experimentally simple; however, the treatment of the data is not easy, although it is now well established thanks to the efforts of the past two decades (Cox and Isham, Smith). For a metrologist the availability of an alternative approach is important, as it is often the only way to detect subtle effects which have previously been overlooked. To be useful, a new method should have as little as possible in common with the traditional approach and it should be of comparable precision. In practice these requirements are difficult to fulfil.

In recent years two completely new methods have been developed, known as "Selective sampling" and the "Correlation method". These have little in common except that they both avoid the need to measure coincidences, the source of most complications. As neither method requires the two emitted radiations to be strictly simultaneous, both can be used for isomeric transitions. To some extent, the novel parity method may be looked upon as a simplified version of the correlation technique, for it also takes advantage of the fact that some of the observed pulses are correlated while others are not. This depends on whether or not they come from the same nucleus.

Let us consider the registered events in more detail. For a decay which occurs in two steps, for example by the emission of a beta followed by a gamma, one can (at least in principle) divide the registered events into two classes: those which have an associated partner in the opposite channel, and those which have not. The number of events observed in a small time interval t can be written as

$$B = b + p \quad \text{and} \quad G = g + p$$

for the beta and gamma channels, respectively. In these relationships p is the number of "paired" events occurring in each channel, whereas b and g represent those which have no partner. Because of p, the numbers B and G, observed in the same time interval, are correlated. This is used in the correlation method by forming Cov (B,G) from which it is possible to deduce p. Unfortunately, dead-time problems, solved to first order only, limit the accuracy of this approach.

Another procedure is also possible. From B and G, which are measured a very large number of times, we can form, for each interval t, the sum

$$S = B + G = b + g + 2p \,;$$

This simple relation provides the key for separating paired and unpaired events by modulo 2 counting. If the probability that a random number $k = 0, 1, 2, \ldots$ is odd is called its parity Π, i.e.

$$\Pi = \text{prob}\,(k \text{ odd}) = \sum_{j=0}^{\infty} \text{prob}\,(k = 2j+1)\,,$$

then we see that $\Pi_S = \Pi_{b+g}$, since $\Pi_{2p} = 0$. Hence, the parity of S depends only on the unpaired pulses b and g.

For simplicity, we first ignore dead times. Then the sum of the events of types b and g forms a Poisson process with parameter $\mu = E(b+g)$. It can be shown that for such a process the parity is given by

$$\Pi = \frac{1}{2}(1 - e^{-2\mu}) .$$

This relation is useful only for $\mu \lesssim 1$ since, otherwise, Π is too close to its limiting value 0.5. By a suitable choice of t, however, this condition can always be met. For the measured parity $\Pi_S = \Pi_{exp}$ the mean value of b + g can then be determined. Since B and G are known from direct measurements, this allows us to find p, which is essentially the count rate of the true coincidences. The activity, as for the coincidence method, is then given by $A = BG/pt$.

In reality, the influence of the dead times must be taken into account. The experiment does not change, but the evaluation of $\Pi_{theor} = \Pi_{b+g}$ becomes more complicated. Let us briefly mention the main reasons. By inserting a dead time, the original Poisson process is distorted. The simplest effect is a reduction in the count rate; the corresponding corrections, usually in the form of transmission factors T_β and T_γ, are well known. Another consequence is that the simple Poisson probabilities for the number of counts are changed to more complicated expressions. The parities, therefore, although still defined as before, are described by more complex expressions. As shown elsewhere, the modified parities can be developed as a series in $x = \mu\tau/t$ of the form

$$\Pi = \Pi^{(0)} + x\,\Pi^{(1)} + x^2\,\Pi^{(2)} + \ldots ,$$

where $\Pi^{(0)}$ is the expression given above for a Poisson process. Explicit forms for $\Pi^{(i)}$ are known up to third order and for both types of dead time. These expressions have been verified experimentally. A particularly unfortunate effect of dead times is that they may cause changes in the classification of events. Clearly, an original event of type p can be transformed into b or g if one of the partner events is lost by a dead time. This requires careful analysis and a model which allows the final classes b and g to be considered as resulting from an original Poisson process deformed by a dead time. A viable solution to this problem was found only recently. Finally, since the pulses b and g are analysed together, we also need a formula which allows us to determine the parity of two superimposed processes. This is possible only if they are independent.

In the actual calculation it is necessary to assume a value for p, so the resulting parity Π_{theor} is a function of p. Equality between measured and calculated parities, therefore, holds only for a certain value $p = p_0$, which can be determined numerically. This p_0 is the number of paired events one would expect in the original situation, i.e. before the dead times, for which all processes are still Poissonian. Given the original numbers of betas and gammas in t, specifically $B_0 = B/T_\beta$ and $G_0 = G/T_\gamma$, all quantities required for the evaluation of the activity are known, with

$$A_0 = \frac{B_0\,G_0}{p_0\,t} .$$

What we have outlined here will be described and illustrated in more detail in the oral presentation. As some decisive experimental tests have not yet been performed, we do not know if the new parity method is capable of achieving an accuracy which is comparable to that of the coincidence method. If this is not so, the method may become little more than a curiosity, albeit a particularly interesting one.

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PL3 s.02

FRONTIERS OF HEAVY ELEMENT NUCLEAR AND RADIOCHEMISTRY

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Summary:
Recent results of both aqueous and gas-phase chemical studies of the heaviest elements (Rf, Ha, and Sg) will be discussed. The prospects for extending the known elements beyond Z=112 and for performing chemical studies of elements beyond Sg (106) will be considered.

Key Words:
Transactinides, Rf, Ha, Sg, spontaneous fission, alpha decay, aqueous & gas-phase chemistry,HEVI

ABSTRACT

Recent chemical and nuclear studies of the heaviest elements will be reviewed. The chemical properties of the transactinide elements 104 (rutherfordium, Rf), 105 (hahnium, Ha) and 106 (Sg, seaborgium) have been studied in both the aqueous and gas phase with manual techniques and with the computer-controlled automated systems HEVI, OLGA, ARCA, and SISAK-LISSY. All of the isotopes of these elements have half-lives of about a minute or shorter and must be studied at the accelerators where they are produced. In addition, their production cross sections are nanobarns or less so "atom-at-a-time" chemistry is required. Their chemical properties have been compared with those of their lighter homologs in groups 4, 5, and 6 of the periodic table to try to assess the influence of relativistic effects, and have shown that their properties cannot be simply extrapolated from those of the lighter homologs.

The elements through 112 are now known and recent confirmation of the predicted stabilization due to the deformed nuclear shells at Z=108 and N=162 make it appear likely that isotopes of elements heavier than 106 may have half-lives long enough for chemical studies. Plans are being made to try to produce the isotope $^{267}107$ via the ^{249}Bk(^{22}Ne, 4n) reaction which is predicted to have a cross section of about 30 picobarns. This isotope is expected to have a half-life of the order of 10 seconds and should be the heaviest member of group 7 of the periodic table so its chemistry should be similar to that of technetium (43) and rhenium (75). Possibilities for extending chemical studies to even heavier elements will be considered.

Nuclear studies include investigation of the half-life systematics of spontaneous fission and alpha-decay and of reaction mechanisms for production of longer-lived isotopes. Current results for spontaneous fission properties such as fission fragment mass-yield distributions and kinetic energies will be presented. The status of a new, high efficiency instrument, the Berkeley Gas-Filled Spectrometer (BGS), which is under construction at Lawrence Berkeley National Laboratory, will be reviewed. Plans to use the BGS in searches for new longer-lived heavy element isotopes, for elements 113 and 114, and as a pre-separator prior to investigations of chemical properties will be discussed.

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PL4
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ISOTOPIC STUDY OF NATURAL FISSION REACTORS AT OKLO AND BANGOMBE, GABON

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Summary: Isotopic study was performed on natural fission reactor samples from Oklo and Bangombe uranium deposits at Gabon. Geochemical behavior of fission products and characterization of the reactors were estimated from the combination of isotopic data.
Key words: isotope, mass spectrometry, fission, neutron capture

The Oklo and Bangombé uranium ores at the Republic of Gabon are unique natural analogues for radioactive waste disposal in a geological media, because fission reactor zones in the ores have preserved abundant fission products for 2 billion years. Since 1982, seven reactors have been newly discovered in these areas (six in Oklo and one in Bangombé) in addition to previously found ten reactors. The new reactors located in deep underground have been little altered, while former ones have been exposed to more oxidized atmosphere because of the deposition near the surface of ore body. In 1990, CEA (Commissariat a l'Energie Atomique) and CEC (Commission of European Communities) jointly started a research program of the newly discovered reactors at Oklo and Bangombé

There are many factors that control the ability of natural reactors to sustain a fission chain reaction. One of the typical geochemical feature of Oklo and Bangombe uranium deposits is that contents of neutron absorption elements such as rare earth elements are extremely lower than other common uranium deposits. Moreover, sufficient amount of natural water existed around these sedimentary uranium deposits. Water might have played an important role to moderate the energy of fission-released neutrons in the reactor. The first neutrons might be released from spontaneous fission of ^{235}U and then be effectively multiplied under the absence of neutron absorbers and the presence of neutron energy moderator. Nuclear reaction cycle possibly occurred in natural reactors is shown in the figure.

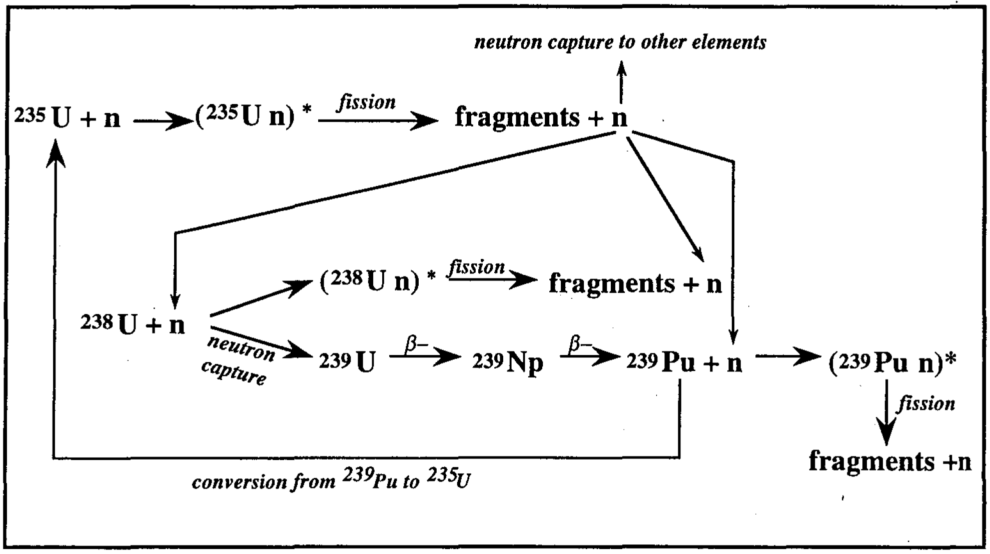

Fig. Scheme of possible nuclear reaction cycle in natural reactors

Many elements in reactors show isotopic anomalies derived from fission and neutron capture reactions. Isotopic analyses of uraninites and some minerals in the natural fission reactors provide a useful information for the geochemical behavior of fission products and characterization of the reactors. Cooperative isotopic measurements by whole rock analysis with inductively coupled plasma mass spectrometry (ICP-MS) and thermal ionization mass spectrometry (TIMS) and by in-situ analysis with secondary ion mass spectrometry (SIMS) make it possible to clarify the migration processes of fissiogenic nuclides over wide range from μm to m.

Fission products in the natural reactors are largely classified into the following four types by the geochemical properties:

1. Compatible elements in the UO_2 and homogeneously distributed in UO_2 matrix: Y, Zr and rare earth elements,
2. Formation of metallic aggregates: Ru, Tc, Rh, Pd and Te,
3. Little compatible elements in UO_2 and largely fractionated from UO_2 matrix: Rb, Sr and Ba,
4. Volatile elements: Cd, Cs and noble gases (Kr and Xe).

These results obtained from the isotopic measurements of natural reactors are in good agreement with those from the microscope observation of artificially irradiated spent fuel.

It is of interest to investigate the trace of long-lived radio precursor like ^{90}Sr ($t_{1/2}$=29.1 yr), ^{99}Tc (2.1x10^5 yr), ^{107}Pd (6.5x10^6 yr), ^{126}Sn (1x10^5 yr), ^{135}Cs (2.3x10^6 yr) and ^{137}Cs (30 yr). Such fissiogenic nuclides cannot be directly detected at present, because these nuclides completely decayed to the stable ^{90}Zr, ^{99}Ru, ^{107}Ag, ^{126}Te, ^{135}Ba and ^{137}Ba, respectively. Their behavior, however, can be estimated from the isotopic deviations (excess or depletion) of their decay products, due to the chemical fractionation between the precursors and their end-products.

Table Nuclear parameters of some natural fission reactors

sample	SC55	KN245	SF84	SD37	BA145
reactor zone	3 (Oklo)	5 (Oklo)	10 (Oklo)	13 (Oklo)	Bangombe
U content (wt.%)	48.0	50.7	24.3	59.4	45.0
$^{235}U/^{238}U$*	0.006940	0.006703	0.005069	0.004630	0.006616
restitution factor of ^{235}U	0.45	0.38	0.304	0.111	0.484
fission proportion					
^{235}U	0.945	0.950	0.906	0.749	0.923
^{238}U	0.025	0.025	0.051	0.179	0.042
^{239}Pu	0.030	0.025	0.043	0.072	0.035
critical duration (x10^5 yr)	2.00	1.70	1.43	0.242	1.10
neutron fluence (x10^{21} n/cm^2)	0.12	0.20	0.798	0.780	0.308
reaction temperature (°C)	290	360	380		

*terrestrial standard value of $^{235}U/^{238}U$ is 0.007252

Determination of nuclear parameters is another main purpose for Oklo study. Isotopic compositions of some elements disturbed by nuclear reactions are useful to clarify the Oklo phenomenon. The natural fission reactors are characterized by the following nuclear parameters; depletion degree of ^{235}U, neutron fluence (n/cm^2), restitution factor of ^{235}U from ^{239}Pu, fission proportion of ^{235}U, ^{238}U and ^{239}Pu, critical duration of reactor, average temperature of reactor. These parameters can be estimated from isotopic measurements of Ru, Pd, Nd, Sm, Lu and U. The estimation values of some reactors are summarized in Table. As shown in Table, fission condition was different in each natural reactor.

Compiling the isotopic results helps a better understanding of the Oklo phenomenon and provides an important prop to the concept of long-term storage of radioactive wastes in geological media.

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FRONTIERS OF ENVIRONMENTAL RADIOACTIVITY

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Summary: The discipline of radioecology encompasses the ecological dynamics of radionuclides and the impacts of environmental radioactivity. Cycling of nuclides, measurement challenges, and predictive abilities are highlighted as frontiers of radioecology.
Key Words: environmental radioactivity, radioecology, radionuclide transport, models

The discipline of radioecology has evolved over the past 50 years into a well-defined mix of ecology and health physics. The discipline has become particularly important as we strive to solve problems such as the Chernobyl accident, disposal of radioactive wastes, and decontamination of land. In this paper, we outline some frontiers of environmental radioactivity, where radioecologists and chemists are required to solve environmental problems. We highlight three topics as major themes of current and future work: cycling of nuclides, measurement challenges, and predictive abilities.

Research on cycling of both stable and radioactive nuclides is an important part of ecology. We focus on radionuclides as potential pollutants, where knowledge of their cycling is needed to estimate impacts on the biosphere, or to make decisions related to management of anthropogenically created radionuclides. These radionuclides not only have a physical radiological half-life, but also have an ecological half-life that defines their persistence within a part of the biosphere. The spatial scale of the cycling is also important since some radionuclides are quite mobile, and can cause concerns at global scales.

Radioecology offers a tremendous opportunity for chemists, especially related to improvements in measurements. Although measurement techniques are constantly improving, radioecologists are pushed to measure lower concentrations; ideally quickly, cheaply and *in situ*. The ability to measure concentrations is central to defining whether an area is "polluted" and some specific problems associated with contamination require unique measurement solutions. We also need techniques to monitor sources of radioactivity such as emissions from a facilities, and we can always improve the ability to overcome background and enhance the signal-to-noise-ratio. This will expand the field of source allocation and footprint analysis, related to our ability to track sources and remediate emissions.

Our third theme is related to predictive abilities. Much of the radioecology work in the present is aimed at estimates of impacts in the future. Therefore, the predictive abilities of radioecologists are of key importance. The corollary of this is the work that is being done on historical dose reconstruction to look at past events and practices. We strive to make predictions that are accurate over very long time-frames. However, stochastic processes make us less confident in predictions as the time-frame lengthens. We usually deal with this through probabilistic analyses or extreme-value scenarios. As the accuracy in predictions improves, radioecology will be a central tool in decision-making to address radiological problems.

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PL6
s.32

NEW FRONTIERS OF DIAGNOSTIC RADIOLOGY

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Summary: There have been many new imaging techniques being developed for the assessments of the various organs and its diseases. In the near future, MRI will become one of the most important modalities in the evaluation of the disease processes, while CT, nuclear medicine and other diagnostic techniques may become complementary to MRI. In any event the newly developing techniques will be refined to non-invasive, less costly, easy to perform and with higher diagnostic accuracy. In the next 10 years, these new modalities will be used more widely in clinical imaging.

Key Words: Diagnostic Radiology, MR imaging, CT, Nuclear Medicine, Ultrasonography

Introduction

There have been many new advances in diagnostic radiology or medical imaging for the past 10 to 15 years. The most prominent and important advances have been observed in Magnetic Resonance Imaging (MRI) since the first clinical introduction in 1980. Computed Tomography (CT) was introduced into clinical practice in 1972, but the new technique of helical CT, recently developed, has made a considerable impact in clinical imaging. There have been also new developments observed in nuclear medicine and ultrasonography. With developments of new imaging techniques, clinical imaging has been performed non-invasively with lower cost, while 3D imaging based on the new techniques has made the interpretation considerably easy with improvement of diagnostic accuracy.

In this presentation, the most recent advances in medical imaging will be discussed with particular attention to MRI, CT, nuclear medicine and ultrasonography.

MR imaging

In order to obtain high quality images in a short imaging time, many techniques have been developed including shorting of TR, reduction of phase encording steps, and reduction of number of acquisitons. Recent techniques involves utilization of Half Fourier technique by using half the data and reconstruction of images by obtaining multiple echoes within the same repetition time. These new techniques have been called RARE, HASTE and echo planar imaging. Although there is a trade-off among temporal resolution, contrast resolution and spatial resolution, it has become possible to obtain images in a very short time.

After fast imaging has become possible, various new imaging studies can be performed. Dynamic MRI can demonstrate the flow pattern in the normal and diseased tissue, whereas MR angiography can demonstrate blood vessels without use of contrast media. Perfusion with use of contrast media can be used for visualization of microvasculature in the tissue, whereas diffusion can demonstrate Brownian motion of proton in the tissue. All these new imaging techniques help to diagnose diseases non-invasively and at an early stage of the disease.

CT

In 1989, helical or spiral CT has been developed and has added a new dimension to the imaging diagnosis with CT. The principle of helical CT consists of continuous rotation of x-ray tube and detector assemblies with continuous radiation from the x-ray tube, while the patient table is continuously transported through the gantry. With this technique, volumetric data can be obtained and, in turn, reconstructed into various images by using SSD, MPR and MIP display techniques. With these techniques, vascular structures and diseases can be demonstrated in 3 dimension, including aneurysms, arteriovenous malformations and steno-occlusive diseases. Furthermore, endoscopic view of the vessels can be obtained.

In addition, the 3D data or volumetric data can be reconstructed as a virtual and endoscopic CT images and internal surface of the organs can be visualized just like an endoscopic study. The virtual CT endoscopy can be applied to the nasopharynx, oropharynx, tracheobronchial tree, stomach, colon

and other hollow viscus such as the gall bladder, the urinary bladder and other air containing structures. 3D reconstructed images can visualize the skin surface as well.
All these new techniques can contribute to non-invasive and accurate diagnosis of the disease processes.

Nuclear Medicine
In nuclear medicine, SPECT systems with multiple detectors and new radiopharmaceuticals have been developed. These new techniques have been applied to the diagnosis of the nervous system, heart, lung, liver, kidney and tumors. In particular, functional images, including cerebral blood flow and cerebral metabolism, have become an essential part of diagnostic work up of nervous diseases. Newly developed I-123 iomazenil is a benzodiazepine neuroreceptor. In nuclear cardiology, many new radiopharmaceuticals have been developed, including Tc-99m MIBI and Tc-99m tetrofosmin for imaging of the myocardium as well as I-123 MIBG and I-123 BMIPP for visualization of the sympathetic system as well as fat metabolism. These new pharmaceuticals have been widely used clinically. The role of each pharmaceutical has to be established.

Ultrasonography
A new imaging technique of the blood vessels has been developed recently on the basis of the mean doppler frequency shift and the integrated Doppler power spectrum. With this technique, background noise is uniformly lower, compared with conventional color Doppler. This new technique is independent of the insonation angle and is much less angle dependent than color Doppler sonography. In addition, the new technique is not subject to aliazing. Because of these advantages, the new technique is now being used for visualization of blood vessels of various parts of the body, non-invasively, simply and for accurate diagnosis.

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PL7
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MÖSSBAUER SPECRTOSCOPY - A POWERFUL NUCLEAR TOOL IN SOLID STATE RESEARCH

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Summary :

Key words : Mössbauer effect, Hyperfine interactions, Mössbauer parameters

A brief introduction into „Recoilless Nuclear Resonance Fluorescence“, called Mössbauer effect, will be presented, followed by a discussion of the main hyperfine interactions and the pertinent Mössbauer parameters. The information content of a Mössbauer spectrum regarding valence state, molecular symmetry, and magnetic properties will be exemplified with typical examples. Selected applications to problems of fundamental solid state chemistry and physics as well as practical applications in industrial research will be discussed to demonstrate the powerful tool of Mössbauer spectroscopy in materials science.

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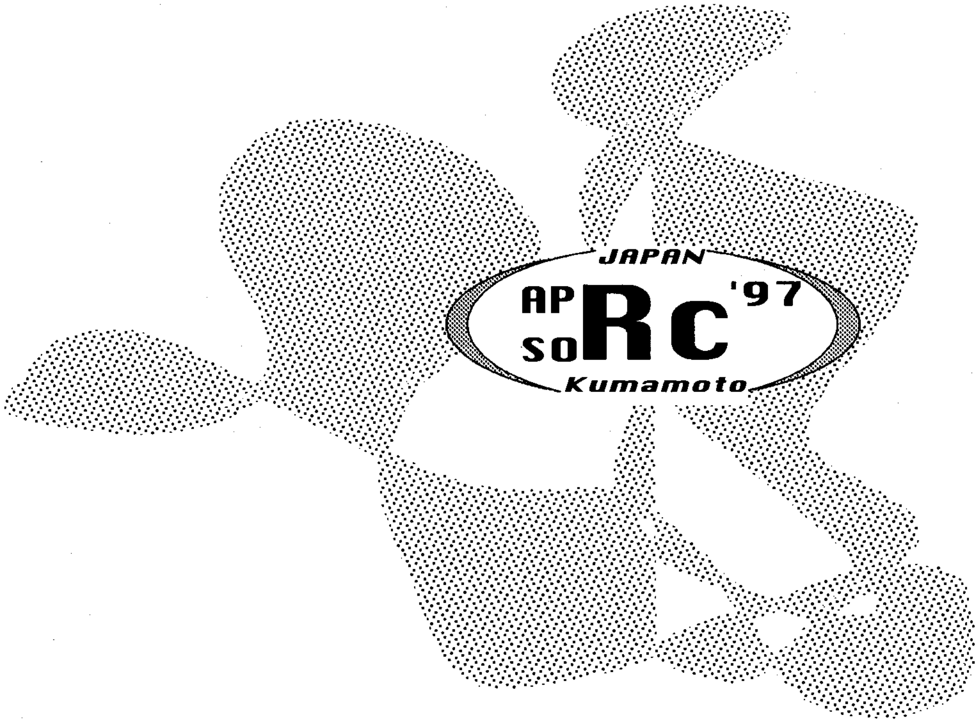
APSORC '97 LIST OF ABSTRACTS

LEAD LECTURES



- KEYNOTE THEME -

EVER ONWARD TOWARDS THE FRONTIERS OF
RADIOCHEMISTRY IN THE
SECOND CENTURY OF RADIOACTIVITY DISCOVERY

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A Retrospective View of a Century of Radiochemistry: Its Growth and Development as a Unique Scientific Discipline

by

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(Paper submitted for presentation to APSORC '97, Kumamoto, Japan; Oct.,1997)

ABSTRACT

This APSORC (Asia-Pacific Symposium) marks the entry into the 2nd century of Radiochemistry and also coincides with the 100th anniversary of Prof. Kenjiro Kimura's birth. Unquestionably, he was a great pioneer in radiochemistry who both experienced and contributed to, a vast growth and development of the field during his lifetime. Many radiochemists in Japan, and in other countries, are indebted to him and his contributions to their careers as a teacher, mentor and effective promoter and supporter of nuclear research.

This paper traces some of the significant highlights in the emergence of radiochemistry from its beginnings up to the unique discipline it has become today characterized by its breadth of scope, its sophistication and its ability to make possible breakthroughs in other disciplines to which radiochemistry has been applied - a range that encompasses the entire spectrum of the physical and life sciences, both pure and applied. Synergistically, these fields have advanced through the deployment of radiochemistry while at the same time radiochemical methods have developed to meet the demands of such applications. Although, in this paper, the emergence of various subfields of radiochemistry, such as radiotracers, radiolabelling, studies of nuclear reactions and synthesis and isolation of artificial radioelements, fission, nuclear geology, etc. are traced starting with radiochemical pioneers like the Curies and Soddy, however some emphasis is given in particular to the development of radioanalytical chemistry during the last half of the century (in the author's experiences in the field).

Examples are presented of the advancement of radioanalytical techniques and their application , particularly to elucidate trace element phenomena in the natural and life sciences, in the study of materials, criminology, space exploration, environmental problems such as Hg, Cd, Pb pollution, monitoring of environmental health hazards through the use of biomonitors like hair, and trace element characterization and tracing of pollutants. Some recent INAA results are also included of novel activable tracer coding for use with specialized and high security materials.

LL2
s.10

FORMATION AND SEPARATION OF CARRIER-FREE RADIONUCLIDES OF FIRST TRANSITION SERIES ELEMENTS

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"Summary:"- RCPAA procedures for formation of different carrier free radionuclides of First Transition Series Elements by α–particle activation of titanium, iron and copper and their individual separation through LLX with liquid ion exchangers have been developed.

"Key Words:"- RCPAA, α-particle, LLX, liquid ion exchanger, γ-ray spectrometry

ABSTRACT

In recent years, the application of carrier free radionuclides has extensively been increased in almost all branches of scientific and industrial research. In using any radioactive isotope, particularly, for diagnostic and therapeutic purposes in biomedical field or in nuclear and radiochemical studies as radiotracer, the purity of the concerned radionuclide is of prime importance. Thus, individual separation of a pure carrier free radiotracer either from its precursor or from the neighboring radioactive elemental species always constitutes an important problem of study to nuclear and radiochemists.

Amongst the various nuclear techniques generally used for the formation of carrier free radioisotopes, such as charged particle activation, fast neutron activation, mother-daughter generator systems, etc., the charged particle activation procedure is perhaps the most appropriate one. In changed particle activation, the experimental parameters, such as, the nature and energy of the projectiles, choice of the target, etc, can suitably be adjusted as per requirements. Moreover, the charged particle activation products are mostly non-isotopic with the target element.

Application of several radionuclides of the First Transition Series (FTS) elements, e.g., ^{47}Sc, ^{48}V, ^{51}Cr, ^{52}Mn, ^{58}Co, ^{57}Ni, ^{65}Zn, etc., in different fields of science and technology, because of their favorable nuclear characteristics, are well established. In producing the desired radionuclides of the FTS, some selected elements, namely, titanium, iron and copper, of specpure quality have been irradiated separately with α-particle beam of appropriate energy varying from 40-50 MeV with beam current of ~ 1-2 μA for a reasonable period at Variable Energy Cyclotron Centre, Calcutta, India. Activation of thick solid targets (metal foils) with the charged particle as projectiles help in the formation of different carrier free radioisotopes of almost all the elements of FTS, such as, ^{47}Sc, ^{48}V and ^{48,49,51}Cr in titanium, ^{52,56}Mn, ^{55,56,58}Co and ^{56,57}Ni in iron and ^{65}Zn and ^{66,67,68}Ga in copper, at degraded energy levels of the projectiles in the irradiated matrices through the nuclear reactions, (α, αpxn), (α, αxn), (α,pxn), (α, xn), etc. having the threshold values less than the beam energy. After irradiation, the activated samples are allowed to cool for successive non-destructive γ-spectroscopic studies and also for their subsequent chemical

Table - I

Radionuclides of the First Transition Series Elements Induced by α-particle Activation of Titanium, Iron and Copper

Target element with isotopic abundance (%)	Radionuclides detected in activated matrix	Half life	Probable nuclear reaction	Threshold energy, E, MeV (approx.)
Titanium	^{47}Sc	3.3 d	(α,αp)	13 - 33
^{46}Ti(7.93)			(α,αpxn)	
^{47}Ti(7.28)				
^{48}Ti(73.94)	^{48}V	16.0 d	(α,pxn)	14 - 36
^{49}Ti(5.51)				
	^{48}Cr	21.6 h	(α,xn)	3 - 27
	^{49}Cr	42.1 m	"	
	^{51}Cr,	27.7 d	"	
Iron	^{52}Fe	8.3 h	(α,αxn)	26
^{54}Fe(5.82)				
^{56}Fe(91.66)	^{52}Mn	5.6 d	(α,αpxn)	22 - 34
^{57}Fe(2.19)	^{54}Mn	312.2 d	"	
^{58}Fe(0.33)	^{56}Mn	2.6 h	(α,3pxn)	
	^{55}Co	17.5 h	(α,pxn)	2 - 36
	^{56}Co	77.7 d	"	
	^{57}Co	271.8 d	"	
	^{58}Co	70.9 d	"	
	^{56}Ni	6.1 d	(α,xn)	7 - 40
	^{57}Ni	1.5 d	"	
Copper	^{61}Cu	3.4 h	(α,αxn)	11 - 22
^{63}Cu(69.09)	^{64}Cu	12.7 h	(α,2pxn)	
^{65}Cu(30.91)	^{67}Cu	2.6 d	(α,2p)	
	^{65}Zn	244.1 d	(α,αpxn)	33
	^{66}Ga	9.5 h	(α,xn)	7 - 27
	^{67}Ga	3.3 d	"	
	^{68}Ga	1.1 h	"	

processing. The radionuclides produced in the irradiated target matrices, as presented in Table-I, have been characterized by their corresponding γ energies and decay studies.

Amongst the chemical separation procedures generally applied to radiochemistry, the technique of solvent extraction, more precisely, liquid-liquid extraction (LLX), is found to be highly effective in the separation of carrier free radionuclides. In destructive analysis of the α-particle activated samples of titanium, iron and copper, suitable radioanalytical methodologies involving LLX have been developed. The efficiency of the liquid ion exchangers, namely, di-2(ethylhexyl) phosphoric acid (HDEHP), tri-n-octylamine (TOA) and triisooctylamine (TIOA) have been fruitfully utilized as extractants in developing the LLX procedures for effective separation of the α-particle induced radionuclides in their carrier-free states. The principle of LLX separation lies in the fact that all the elements of FTS are very much susceptible to form various types of complexes, both cationic and anionic, in aqueous acidic and alkaline media under specified experimental conditions. In some cases, presence of oxalic acid, acetic acid or H_2O_2, help in complexation of the elemental species resulting in an enhancement of separation of the carrier free radionuclides. The applicability of each of the separation procedures is very much dependent on the nature of the acid and its acidity or the alkalinity in the aqueous medium and also on the concentration of the liquid ion-exchangers in the extraction systems. The extractant solutions at different concentrations have been prepared by adding calculated amounts of cyclohexane as diluent to the reagents.

The extraction separation of the bulk target matrices, namely, iron and copper, have been studied radiometrically using ^{52}F(8.3 h) and ^{64}Cu(12.7 h) formed by the respective nuclear reactions, $^{54}Fe(\alpha,\alpha 2n)^{52}Fe$ and $^{65}Cu(\alpha,\alpha n)^{64}Cu$, respectively as radioindicators for the corresponding elements. However, in case of titanium, since no suitable γ-emitting radionuclides is produced in the matrix under the experimental conditions, verification for extraction separation of the elements has been performed spectrophotometrically. The purity and the extent of extraction-separation of the carrier free radioisotopes in different phases have been verified by taking recourse to γ-ray spectroscopic studies.

Thus, the developed RCPAA procedures, because of their efficiency in producing and separation of different important carrier free radionuclides of FTS elements, may have potential applications in biomedical and industrial research.

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LL3
s.22

FRONTIERS OF POSITRON AND POSITRONIUM CHEMISTRY IN CONDENSED MEDIA

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Summary: Our work proves that the positron annihilation spectroscopy is an excellent tool to follow the structural changes in chemical species. We present four examples to support the above statement.

Key words: conformal structural transformation, defect structure, electrodeposited coatings, lifetime distribution, ortho-para conversion, polymers, positron lifetime spectroscopy, solid state NMR, spin-crossover

1. Spin-crossover

For complexes of Fe^{II}, where the central Fe^{II} undergoes the temperature induced high-spin → low-spin transition $t_{2g}^4 e_g^2 \rightarrow t_{2g}^6$ $\left({}^3T_{2g} \rightarrow {}^1A_{1g}\right)$ [1], the *ortho*-positronium lifetime detects sensitively the spin transition, as it can be seen for the compound $[Fe(1\text{-ethyl-}1\mathit{H}\text{-tetrazole})_6](BF_4)_2$ (Figure 1). At temperatures above the spin transition ($T > T_c$) where the Fe^{II} ions are in the high-spin state, the *ortho*-positronium lifetime is significantly lower than at $T < T_c$ (low-spin state), since the unpaired electron spins induce ortho-para conversion.

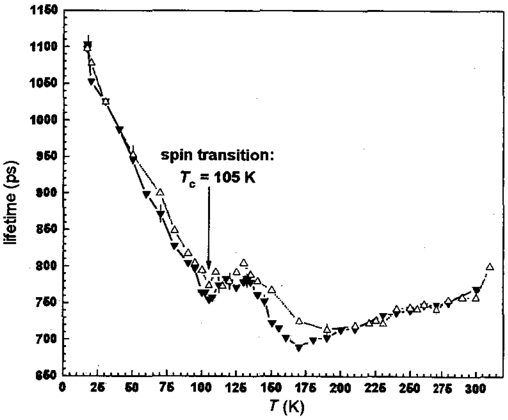

Figure 1 *Ortho*-positronium lifetime for $[Fe(1\text{-ethyl-}1\mathit{H}\text{-tetrazole})_6](BF_4)_2$. Open symbols: heating direction, full symbols: cooling direction.

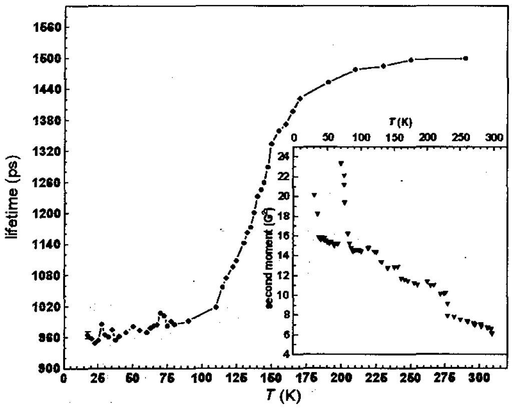

Figure 2 *Ortho*-positronium lifetime for $[Zn(1\text{-propyl-}1\mathit{H}\text{-tetrazole})_6](BF_4)_2$. Insert: Second moment of solid state ^1H NMR spectra.

2. Conformal structural transformation

For the compund $[Zn(1\text{-propyl-}1\mathit{H}\text{-tetrazole})_6](BF_4)_2$ we found that the *ortho*-positronium lifetime ($\tau_{o\text{-}Ps}$) displays a large step between 175 and 100 K (Figure 2) [2], the interpretation of which was possible on the basis of solid state ^1H NMR measurements (see insert). The temperature dependence of the second moment (M_2) of the NMR spectra, which is proportional to the average proton-proton distance, showed that at $70 < T < 90$ K there occurs a structural change while which the molecular motions are somewhat ceased. At 90 K, the propyl sidechains start to move more freely (M_2 decreases). Possibly, this change triggers the abrupt increase in $\tau_{o\text{-}Ps}$. So, we can conclude that the *ortho*-positronium lifetime detects even slight changes of the molecular structure which does not effect the crystal symmetry and the cell parameters.

3. Defect structure

Electrodeposited coatings contain large amount of void type crystal defects and these defects affect the protective features of the layer. The conditions of the preparation, however, modify the defect structure of the layer very much. In Figure 3 we present an example of Cr coatings on Cu prepared using different current densities.

At 40 A.dm^{-2} the two observed positron lifetimes indicate two types of crystal defects: monovacancies (τ=170 ps) and voids (τ=325 ps). At this current density monovacancies dominates (I_1=75%). When larger current densities are applied the monovacancies aggregate and form small vacancy clusters (τ=220 ps) while the size of the large clusters increases indicated by the increasing lifetime value. At the same time the relative weight of large voids increases from 25% to 65%.

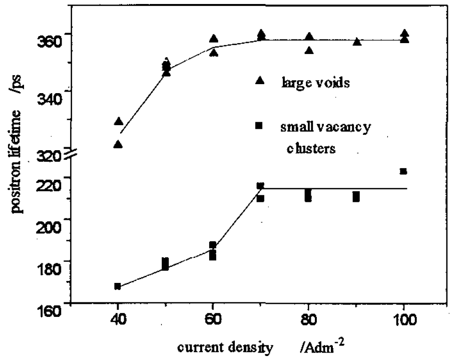

Figure 3: The lifetime parameters of the two positron states observed in Cr coatings

4. Free volume distribution in polymers

Free volumes larger than vacancies are important specieses determining the properties of polymers. In this case the lifetime distribution of o-Ps atoms may provide valuable information on the amount and the size distribution of free volume holes [3].

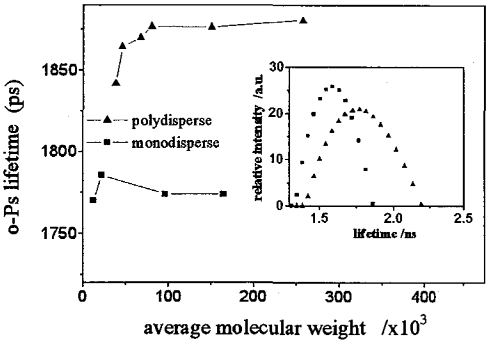

Figure 4: The average o-Ps lifetime and its distribution in mono and polydisperse poly-(methylmetacrylate) samples. The distributions are given for samples of $M_n \approx 100000$ in both cases.

Figure 4 presents the results of a recent study in which the effects of molecular weight and dispersity were investigated in poly-(methylmetacrylate) (PMMA) samples. The o-Ps formation intensity was around 23% in both monodisperse and polydisperse samples, so, the only difference between the two sets of PMMA was the size distribution of the samples.

In the case of polydisperse PMMA samples the larger average o-Ps lifetime indicates that the average size of free volume holes is larger than in the case of monodisperse samples. This difference is emphasize by lifetime distribution curves (see insert). Due to the free volume model of o-Ps annihilation these lifetime distribution curves represents the size distribution of free volume holes, as well.

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LL4
s.52

UNDERSTANDING OF BURNING RATE BEHAVIOUR BASED ON NEUTRON ACTIVATION ANALYSIS OF ROCKET PROPELLANT INGREDIENTS

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Summary : Neutron activation analysis of the ingredient Al (~ 17%) present in a rocket propellant grain showed a variation from 16 to 21%. Different trends for Al content and burning rate of activated samples were observed over different portions of the grain.

Key Words : Rocket propellant, Neutron activation analysis, Al content, Burning rate.

A solid rocket propellant can be non-uniform in its structure and composition because of its large size and therefore may exhibit an appreciable variation in its performance parameters such as burning rate, calorimetric value, specific impulse, etc. In a composite propellant such variations may further magnify due to distinct difference in the density of the ingredients. It is imperative to understand the irregularity in the distribution of materials in a propellant grain and more importantly its causes and effects on the behaviour of the propellant grain, so as to arrive at the appropriate remedial measures. It has been reported that non-uniformity in the propellant density and cal-val in a large-size grain is related to viscosity and temperature of the final mix. Variation in the mechanical properties with respect to different locations in a grain has been interpreted on the basisi of heat transfer. There are several attempts for evaluating homogeneity of the propellant grain, however, all these have been on a macroscopic scale. This work deals with microscopic evaluation of the material distribution inside a large propellant grain using the instrumental neutron activation analysis technique. A correlation is also sought between burning rate behaviour of small pieces of the large-size grain and their aluminium content.

A typical composite propellant formulation containing the major ingredients as NH_4ClO_4 (65.5%), Al(17%), Fe_2O_3(1%) and polymeric binder (13.5%), that was cast under vacuum and cured by thermal treatment at 65 ± 5 °C was used. The cured tubular block of size 110 mm OD and 150 mm ht. was cut transversely into three equal size portions and each portion in turn was cut into pieces of equal dimensions (8 mm x 8 mm x 40 mm). A few of these were selected to cut into two equal halves and each half was evaluated for burning rate under nitrogen atmosphere with a pressure of 6.8 MPa using a standard strand-burner system coupled to an acoustic unit. The remaining samples were neutron activated for 15.0 min. using a ^{252}Cf source (flux : 10^9 n s^{-1}). After a 40 s time lapse, ^{28}Al activity in the sample was measured using an HPGe detector coupled to a 4 k MCA. Same(8X8X40 mm^3) size standards were prepared by casting and curing thoroughly mixed slurries containing the weighed amounts of Al, NH_4ClO_4, Fe_2O_3 and the binder.

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Burning rate was determined in a large number of propellant pieces which represented the complete grain. The values obtained for this rate are seen to have a wide spread over a range of 10 - 14 mm s^{-1} having a maximum around 11 mm s^{-1}. Using specially prepared standard samples having a typical propellant composition (NH_4ClO_4 : 66%, Al : 19%, Fe_2O_3 : 1% and binder : 14 %), the deviation in burning rate was found to be less than ± 0.2 mm s^{-1}. This deviation is well within the reported value. In order to understand the cause for this burning rate variation, samples from the same propellant grain were estimated by neutron activation analysis technique for their Al content. For this, a standard calibration curve was obtained after carefuly weighing Al powder and other ingredients, mixing them thoroughly in a weighed quantity of the polymer matrix. The mix was then cast, cured by the conventional way and finally cut into 8X8X40 mm^3 size. The plot of observed activity versus the Al content was found to be a straight line, the slope of which matched with that of the plot wherein the content was determined by the usual chemical procedure.

Al content in the sample pieces of the entire grain, determined using the calibration curve, was found to be varying over the range of 16-21%; here the standard deviation was found to be less than ± 1.5%. These data presented in Fig. 1 show microscopic radial distribution of Al over the top, middle and bottom portions of the grain. It is seen that at any particular radial angle there is a clear cut difference in the Al content between the inner and outer regions of the bottom portion. The Al content is less than 19% over the inner region and greater than this value over the outer region. No such separation is seen between the inner and outer regions of the middle and top portions of the grain. Regarding the standard deviation, it is larger (18.7 ± 1.7 %) in the bottom portion as compared to that in the middle (17.2 ± 0.9%) and top (17.6 ± 0.8%) portions. Higher average value of Al content in the bottom portion indicates that Al settles more in this portion during the curing process.

In order to understand the relationship if any, between Al content and burning rate, neutron irradiated pieces were evaluated for burning rate. Radial distribution obtained over the inner and outer regions of the top, middle and bottom portions showed wide variation in the rate. Although both the burning rate as well as Al content varried from sample to sample, no correlation could be evolved from these data in the case of top of middle portions of this grain. The data for the bottom portion, however, showed some trends. Over the region where Al content is low, burning rate decreases with increasing Al till it reaches a minimum around 19%. This trend then reverses and the rate increases with further increase in the Al content. The former trend has resemblence with this reported one wherein a 20% decrease in burning rate was observed when the Al content was increased from 16 to 24%.

Reproducibility in the performance of a propellant depends upon uniformity in burning rate throughout the grain. This reproducibility in turn would depend on the uniformity in the distribution of ingredients. The present studies demonstrated that the latter can easily, rapidly and accurately be determined following the neutron activation analysis. Thus the NAA technique can be used effectively as an analytical tool in a propellant / explosive factory. It can help evaluate the process parameters at any stage of mixing, casting and curing.

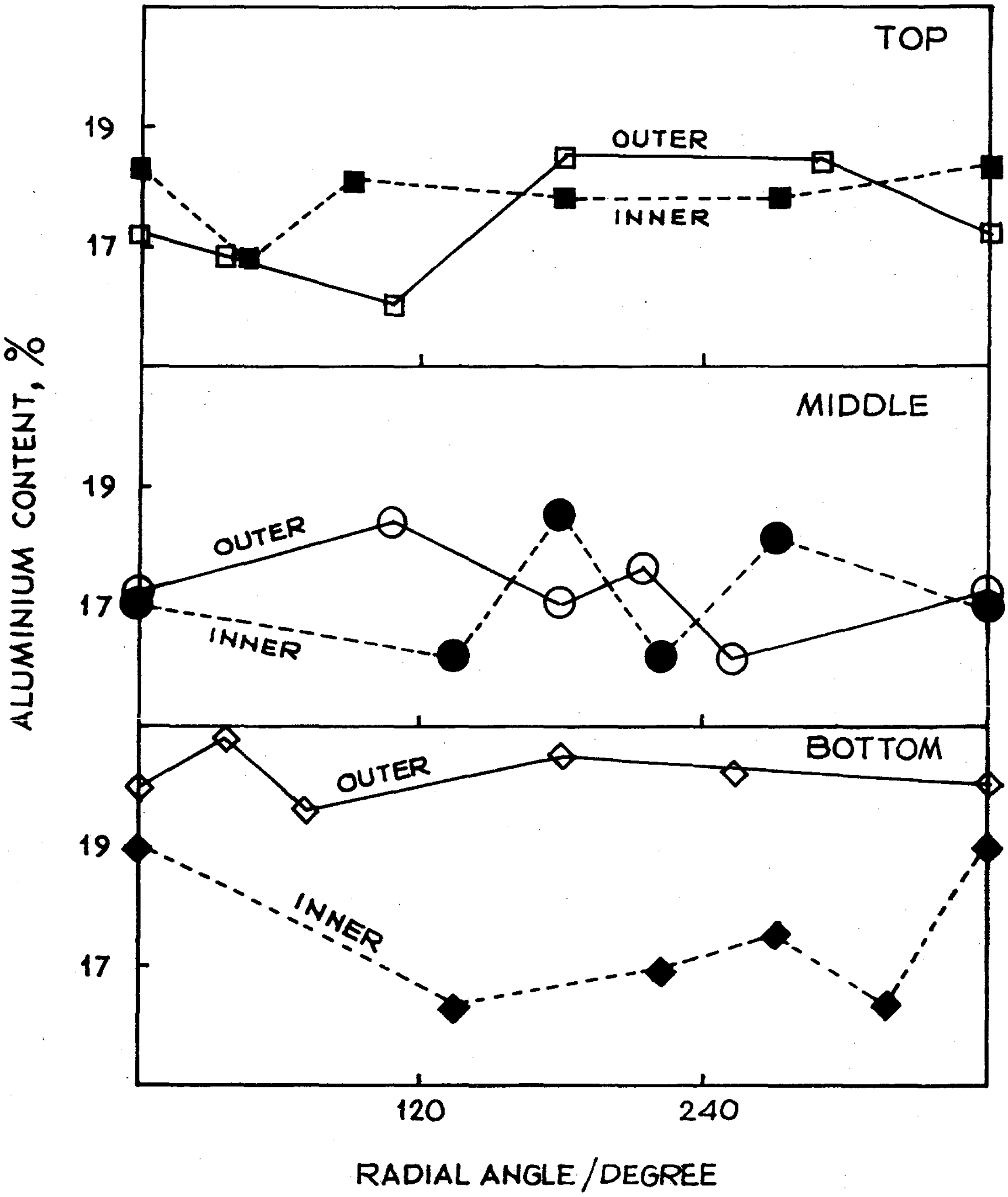

FIG. 1 - ALUMINIUM CONTENT AS A FUNCTION OF RADIAL ANGLE

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IN-VIVO NEUTRON ACTIVATION ANALYSIS FOR MEDICAL APPLICATIONS.

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Summary: This paper describes the design and uses of in-vivo activation analysis for the determination of total body calcium and total body nitrogen of humans. The data is useful in a variety of clinical situations dealing with the diagnosis and management of patients with osteopenia, nutritional deficiency, renal dysfunction, transplantation, cancer, thyroid dysfunction and many others.
Key Words: in-vivo activation analysis, total body calcium, total body nitrogen.

Total Body Calcium: Almost all the calcium in the human body is in the bone and therefore the measurement of total body calcium is an indicator of the bone mineral status of an individual. When diseases occur that results in the disturbance of calcium metabolism, it often manifests itself in the form of changes in the total body calcium. Examples of such diseases are many and includes the common women's disease 'osteoporosis' which affects over 25% of post menopausal women.

An effective non-invasive method of measurement of total body calcium is by in-vivo neutron activation analysis. Our facility is based on the thermal neutron reaction $^{48}Ca(n,\gamma)^{49}Ca$. The neutron sources consist of 10 Curie Pu-Be sources making a total of 110 Curies. These sources are placed in such a way that an area of 60 in. x 40 in. of the patient, from the sternal notch to mid thigh, is irradiated with a uniform flux of neutrons. The neutrons are thermalised essentially by the body water leading to the above thermal neutron activation reaction. The irradiation is done for 20 minutes and the resulting 10 MeV gamma rays from the 3.3. minute ^{49}Ca induced is counted in a shielded whole body counter for another 20 minutes. The amount of ^{49}Ca induced in an individual is a measure of the total body calcium. It is necessary to determine whether this measured total body calcium is normal or not. We have measured a large number of normal individuals and have developed a mathematical formula to calculate the normal total body calcium on the basis of the height of the individual. Thus if the measured total body calcium in a patient is divided by the calculated normal value, we then get the Calcium bone index, CaBI, which is used as a clinical indicator of osteopenia.

Total Body Nitrogen: The measurement of total body nitrogen is an indicator of the protein status of an individual. This, especially in the case of a patient undergoing treatment, indicates the general health of the patient. For example, when a person with renal dysfunction or cancer is under treatment, the apparent gain in weight may not be of any significance, if it is only due to an increase in body water and not due to the production of protein or muscle mass.

Total Body Nitrogen measurement is based on the nuclear reaction $^{14}N(n,\gamma)^{15}N$. Since ^{15}N has a half-life of a fraction of a second, the irradiation and counting have to be done simultaneously. This poses the problem of large background signal from the neutron source itself. In our facility ^{252}Cf is used as a neutron source since the signal to noise ratio is better in this case than the Pu-Be source. The disadvantage, however, is that the ^{252}Cf source has a 2.2 year half life only compared to 85 years for the Pu-Be source. Thus the former source has to be replaced every four years or so and this adds to the cost. Prompt gamma activation signal is affected by the body size and shape. In order to correct for this, in stead of using the ^{15}N signal (10.8 MeV) as the indicator of nitrogen status, the nitrogen to hydrogen ratio is used. For this purpose the hydrogen content of humans is taken to be 10% of body weight. Hydrogen undergoes the prompt gamma reaction $^{1}H(n,\gamma)^{2}H$ yielding a 2.2. MeV signal. The ratio of the 2.8 MeV to 10.8 MeV signal is used in the measurement. As in the case of calcium, we have measured a large number of normal persons for their total body nitrogen content and have derived a mathematical relationship between the body size and the total body nitrogen. Thus we are able to calculate the normal total body nitrogen of a patient and by knowing the measured amount we can calculate the deviation from normal as a nitrogen index, NI. This nitrogen index is used as a clinical indicator of protein status of a patient.

Both the CaBI and NI have been used by us as clinical indicators in a variety of situation. These are useful as primary tools of diagnosis for medical intervention. The effectiveness of medical intervention can be monitored by periodic measurements of CaBI or NI in individual patients. In addition, a variety of clinical trials uses these techniques. In the area of treatment of osteoporosis, a variety of treatment regimes have been developed. These use agents either to stimulate bone growth or alternatively to suppress resorption. We have done extensive clinical studies using fluoride as the medium of bone growth in osteoporotics. Similarly a number of clinical trials using bisphosphonates, the anti-resorption agents, are being conducted to test newer generation of drugs for the treatment of osteoporosis.

Newer Techniques: In the last few years, techniques based on x-rays have been developed to measure bone mineral content. These are becoming rapidly popular and in clinical use. This paper will examine the advantages and disadvantages of both the in-vivo neutron activation and the x-ray methods. The former has a larger radiation dose (approximately 3 to 5 mSv) compared to the latter (approximately 0.05 mSv). However both the techniques continue to be used in our laboratories since the x-ray method requires the patient to lie flat during the measurement. Many osteoporotics, especially ones with a severe condition, cannot do so and hence in-vivo neutron activation methods is used with them. The results from the two techniques correlate well in most cases with some exceptions. These will be discussed in this paper.

Design and Cost of in-vivo techniques: This paper will examine the design of a hospital facility for in-vivo activation with a view to reduce capital and operating costs while maintaining accuracy and precision. The irradiation facilities can be built in-house with materials commonly available. The whole body counter can also be built in-house. In the measurement of total body calcium, the counting can be done with a shadow shielding thereby reducing the capital cost of building the whole body counter. The data collection and processing can also be simplified by independently operating each detector and summing up the net counts from all the detectors. This avoids the necessity of expensive spectrum stabilizers if all the detectors are routed through the same amplifier system.

Our experience with these in-vivo systems in the diagnosis, monitoring and management of patients with a variety of diseases will be discussed. Their use in clinical trials for the development of new drugs will also be examined.

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TRACE ELEMENTS IN BONE DETERMINED BY NEUTRON ACTIVATION ANALYSIS: ITS ROLE IN OSTEOPOROSIS

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Summary

There is an evidence that some of the essential trace elements are crucial determinants of bone health. Up to 25 trace elements in iliac crest of the apparently health human subjects were determined by INAA and other techniques and their relations were discussed.

Key words:

Trace elements in bone, Bone mineral density, neutron activation analysis, osteoporosis

There is still considerable uncertainty about normal mineral metabolism in bone and, in particular, the genesis of the reported geographic variability in bone health and bone mineral density, BMD. There appear to be major discrepancies between different countries in terms of BMD and habitual dietary intake. It is believed that calcium, phosphorus and vitamin D are the major determinants of peak bone mass. However, there is now evidence that imbalance diets in general, which may lead to poor nutritional status with respect to several essential nutrients in particular, Mg, K, Zn, Cu, Mn, F and vitamins C and K and few other trace elements, are also crucial determinants of bone health. On the other hand, several elements such as Cd, Al and Pb are known to be deleterious to bone metabolism. Because of the importance of trace elements in bone, we have determined trace elements in bone by INAA and by other techniques. The main techniques that are available for the analysis of bone samples are atomic absorption spectrometry, instrumental neutron activation analysis, inductively coupled plasma optical emission spectrometry, and inductively coupled plasma mass spectrometry. Reference materials for bone are rather scarce. The only two materials currently available are NIST SRM 1486 (bone meal) and SRM 1400 (bone ash). Bone meal is the more appropriate CRM (certified reference material) for the studies proposed, although it lacks certification for three of the elements of primary interest in these studies (i.e. Cu. F and Mn). Among these elements, Cu is determined by GF-AAS, and F is determined by Ion Selective Electrode technique using fluoride electrode.

Experimental

Bone biopsies were obtained from apparently healthy subjects who were undergoing orthopedic surgery due to any other reason than osteoporosis. Here we will present the results of only three males and one female subjects ages between 17 and 36. They filled the Osteoporosis Project Questionnaire that contains information on life style , dietary intakes, and the reason for surgery. Bone mineral density was measured prior graft removal for most of the subjects.

A 3-5 g of bone graft was obtained from the anterior iliac crest using a Ti or a stainless steel osteotome and placed into prewashed polyethylene bags and stored at -20 ^{0}C in a deep freezer. The iliac crest of hip bone is chosen because of local variation in the elemental concentration along the iliac crest is minimal and these biopsies are commonly taken clinically on patients. The biopsies taken were separated in to cortical and trabecular parts with an osteotome. The use of trabecular part of the iliac crest for trace element analysis has the advantage of reflecting rapidly changes in the composition of bone due to external parameters, including medication. The sample was divided into smaller parts and weighed. For the removal of possible surface contamination, both cortical and trabecular parts were placed in a polyethylene bottle containing 50 mL 5% citric acid solution and washed for about 30 seconds in the ultrasonic shaker. Blood was removed by placing the both trabecular and cortical parts of the bone samples in 50 mL 5% glucose solution (isotonic solution) for about 60 minutes in the ultrasonic shaker, repeating the procedure for six times. Fat and muscle was removed placing the sample in 50 mL diethyl ether for about 15 minutes. The bone sample in a polyethylene bag was frozen in liquid nitrogen and then grounded in a Teflon container to produce a homogenous sample. The sample was dried at 60 ^{0}C for several hours until constancy of weight has been achieved. For easy and fast separation, it is recommended that blood, muscle and fat be removed immediately after the bone sample is obtained from the subject.

Bone samples weighing 200 to 300 mg were irradiated with thermal neutrons at Triga Reactor in Istanbul for 3 hours. After about 6 days of cooling, gamma rays coming from the radioactive isotopes were measured. We have also subjected to, both cortical and trabecular parts, short and long irradiation at Massachusetts Institute of Technology , USA. We have used NIST SRM 1486, bone meal, as a standard. The copper was determined by GF-AAS. Table I shows the result of Na, K, Zn, Fe, Cu, Ba, Cr, Se, As, Cd, Br, Hg, Rb, Mo and Sc in trabecular and cortical bone of iliac crest. Besides these elements, most of the rare earths were also determined by INAA and fluorine by ISE using fluoride electrode.

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Table I Contents of some minor and trace elements in cortical(C) and trabecular(T) bones of iliac crest four subjects ages between 17 and 36, determined by INAA and GF-AAS(Cu).

Elements	**2C**	**3C**	**4C**	**5C**	**Ave ± Std**
Na(g/kg)	7.20	2.70	3.50	4.60	**4.50 ± 1.96**
K(g/kg)	2.10	3.30	1.60	5.20	**3.05 ± 1.60**
Fe(mg/kg)	62	180	56	81	**95 ± 58**
Zn(mg/kg)	120	62	55	76	**78 ± 29**
As(mg/kg)	0.27	0.37	0.23	0.52	**0.35 ± 0.13**
Se(mg/kg)	0.77	0.69	0.32	0.77	**0.64 ± 0.22**
Rb(mg/kg)	1.0	1.2	1.1	0.8	**1.0 ± 0.2**
Mo(mg/kg)	0.4	1.2	0.9	1.4	**1.0 ± 0.4**
Cd(mg/kg)	1.70	0.36	0.27	2.00	**1.08 ± 0.90**
Ba(mg/kg)	21	16	13	8	**15 ± 5**
Cr(mg/kg)	1.20	0.90	0.40	0.48	**0.75 ± 0.37**
Br(mg/kg)	2.90	8.10	0.77	0.50	**3.07 ± 3.52**
Cu(mg/kg)	4.22	4.07	3.58	2.27	**3.54 ± 0.89**
Hg(mg/kg)	3.2	1.9	2.2	2.3	**2.4 ± 0.6**
Sc(μg/kg)	6.6	5.3	3.0	2.6	**4.4 ± 1.9**

	2T	**3T**	**4T**	**5T**	**Ave ± Std**
Na(g/kg)	3.70	2.90	3.50	5.00	**3.78 ± 0.8**
K(g/kg)	0.76	1.20	1.20	1.10	**1.07 ± 0.21**
Fe(mg/kg)	34	110	280	37	**115 ± 115**
Zn(mg/kg)	53	53	88	97	**73 ± 23**
As(mg/kg)	0.15	0.23	0.14	0.20	**0.18 ± 0.04**
Se(mg/kg)	0.76	0.40	0.66	0.60	**0.61 ± 0.15**
Rb(mg/kg)	1.6	0.4	1.6	2.8	**1.6 ± 1.0**
Mo(mg/kg)	0.8	1.0	0.8	1.0	**0.9 ± 0.1**
Cd(mg/kg)	0.01	0.08	0.54	0.28	**0.23 ± 0.24**
Ba(mg/kg)	16	19	14	20	**17 ± 3**
Cr(mg/kg)	0.84	0.76	0.58	2.30	**1.12 ± 0.79**
Br(mg/kg)	0.61	0.96	1.60	0.51	**0.92 ± 0.49**
Cu (mg/kg)	4.12	4.18	3.04	2.53	**3.47 ± 0.82**
Hg(mg/kg)	2.1	2.2	1.3	2.3	**2.0 ± 0.5**
Sc (μg/kg)	5.5	4.1	3.0	4.4	**4.3 ± 1.0**

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NUCLEAR ACTIVATION AND RELATED TECHNIQUES FOR ANALYSIS OF HUMAN TISSUES AND DIETARY SAMPLES

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Summary : Neutron activation analysis (NAA) and other techniques have played important roles in the health sciences. NAA has been used for the assay of many trace elements (TE) in foods and human tissues. Results from recent studies are used to reassess total body content and dietary intake of TEs.
Key words: Nuclear techniques, trace elements, foods, tissues, reference materials, Reference Man

The past 2 decades (particularly during the 80s), have witnessed a surge of analytical refinements that addressed the concept of total quality control (e.g. biological and analytical standardization for collection and analysis of biological specimens). A number of analytical techniques (based on both nuclear and non-nuclear methods) have contributed to the growth of trace element data in biological media as documented in numerous sources. For example, problems faced by trace element researchers in their quest to generate reliable analytical results in biological systems, and proposed solutions to overcome those problems have been documented (1). In particular, several sources of potential errors (stemming from the influence of presampling factors) in dealing with biological samples were identified and steps to identify them through biological standardization as a part of of the measurement process was enumerated (2).

As an example of the growing appreciation of the collective capabilities of different techniques, their suitability for analysis of radiologically important trace elements such as Cs, I, Sr, Th and U, in food and tissue matrices is shown (Table 1). This evaluation is the result of experience gained over a long period of applications in various international investigations as summarized (3). The nuclear activation techniques have played a major role by contributing to the determination of a number of trace elements that are difficult to determine by other methods. Simultaneously, the enhanced capabilities of non-invasive analytical techniques such as NAA and X-ray fluorescence (XRF) generated data of exceptional analytical quality (4). Since then extensive programs related to tissue and body fluid analysis have been carried out to establish reliable concentrations particularly for trace elements (e.g. 5, 6). Similarly, well planned investigations on human diets on an international scale have also been carried out to determine the daily intake of minor and trace elements, and reliable data are now available (7). The results are a combination of a stringent analytical procedures used to in conjunction with analysis of appropriate reference materials for quality control. Further, consistent dietary intake and tissue concentration data are emerging for Al, As, B, Cd, Cr, Li, Mo, Si and Sn, among others. Thus, the database has expanded to include elements not included in Reference Man (8), and therefore, a reevaluation effort was undertaken.

Reference concentrations and total body contents: Rather large collection of data was possible for Cu, Fe and Zn, and for Mn in specimens such as hair and liver as discussed (1). For a number of elements ICRP-23 provides total body estimates (8). These are shown in Table 2 along with the *present* assessments, for comparison. The comparison reveals that for Br, Cs, I, Pb and Zn, the average values for the total body content derived from the present evaluation have remained close to the 1975 ICRP-23 estimates. For Al, Ba, Cu, F the present estimates show an increase indicating improvements in analytical methods or increases in dietary intake; presumably due to a combination of both conditions. The cause for reduction of Rb value is not very clear, but also points to refinement in analytical methodology. For As, Cd and Rb, the total contents have declined suggesting improvements in the analytical determination, especially for As and Cd, which occur at low concentrations in most foods, and therefore indicating diminished dietary intake. For B, Co and Mn the uncertainty of the 1975 evaluations have been reduced. For Au and Cr the 1992 estimations are far below the 1975 values clearly indicating analytical problems of earlier assays. Estimates for the total body contents for Ni (4-6.5 mg), Se (14-21 mg) and Si (2000-3000 mg) are new (not reported in ICRP-23), hence should be regarded as *provisional* pending additional investigations to reach a reliable conclusion.

Reference values for daily dietary intake: Examples of the changes taking place in the dietary inorganic analysis data for a few trace elements are presented in Table 3. The ICRP-23 (8) evaluation, which may be regarded as a check point for earlier intake estimations, summarizes the data recorded until the late sixties. Subsequent studies carried out in the U.K. (9) and in Finland (10) are good illustrations of the efforts undertaken in the seventies. These investigations reflect an improved understanding of the analytical techniques applied for food analysis, supported by efforts to evaluate the precision and accuracy of the methodologies by analyzing available reference materials. Thus, several gross over-estimations in earlier studies, e.g., for Al, As, Li and V were exposed. In contrast to the earlier estimates in daily dietary intakes, decreases by factors of 2 for Cu, 2-5 for Cr, 5-10 for Cd, Hg and Pb, and >10 for Ag, As, Co, Li, Sb and V were observed. It is conceivable that the errors associated with the older values were of methodological origin. In specific cases, the decline of total intake can partly be linked to environmental regulations that went to effect (e.g. elimination of Pb from gasolene).

It is concluded that modern analytical techniques, particularly nuclear activation techniques in various versions have been very useful for reevaluating the elemental composition of reference man.

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Table 1. Analytical methods for Cs, I, Sr, Th and U in tissues and foods

Desired analytes	**Foods diets**	**Soft tissues**	**Bone samples**
Cesium	INAA/GF-AAS/FES	INAA/GF-AAS/ICP-MS	INAA
Iodine	ENAA/RNAA/INAA	ENAA/RNAA/INAA	INAA(?)
Strontium	ICP-ES, Flame AAS(s), PIXE, INAA(?)	ICP-ES, RNAA	ICP-ES Flame AAS(s)
Thorium	ICP-MS/RNAA,∝Sp.Metry	ICP-MS/RNAA	RNAA/ICP-MS(?)
Uranium	ICP-MS/RNAA Delayed neutron counting, ∝Sp.Metry	ICP-MS/RNAA Delayed neutron counting	RNAA/ICP-MS(?) Delayed neutron counting

Footnotes: (?) denotes concentration as a factor.
(s) refers to measurement following chemical separation.

Table 2:Impact of reliable analysis on trace elements in Reference Man

Element	**ICRP-23(1975) Average mg**	**Based on recent results Average mg**	**Range mg**
Aluminum	61	92	47.5 - 168
Arsenic	18	2.8	2.6 - 3.4
Barium	22	49	31.5 - 92.5
Boron	<20	14	12.3 - 32
Bromine	200	210	102 - 247
Cadmium	50	19.5	12.6 - 36.4
Cesium	1.5	1.7	1.0 - 2.6
Chromium	<7	0.7	0.5 - 1.65
Cobalt	<1.5	1.4	-
Copper	72	121	88 - 228
Fluorine	2.6	16.3	12 - 30
Gold	<10	<0.3	-
Iodine	13	11.7	7 - 19
Iron	4200	5000	4080 - 5980
Lead	120	138	60 - 380
Manganese	<12	11.4	7 - 20
Nickel	-	(5)[b]	(4 - 6.5)
Rubidium	680	258	(150 - 400)
Selenium	-	(17)	(14 - 21)
Silicon	-	(2500)	(2000 - 3000)
Zinc	2300	2220	1540 - 2300

[a] Data pooled from reviews and interim evaluations mentioned in ref. (14)
[b] Values for Ni, Se and Si are new and preliminary; not given in ref. (8)

Table 3:Impact of reliable analysis on estimations of dietary intake

Element	**ICRP-23 (μg/d)**	**Present estimates[a] (μg/d)**
Arsenic	1000	50-100
Barium	750	600
Boron	1300	1800
Cadmium	150	15-25
Cesium	10	10
Copper	3500	1500
Lead	440	50-100
Lithium	2000	30-100
Silver	70	5-10

[a] Data pooled from references (1,6,7,11-13).

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CHARGE-INDUCED X-RAYS (CHIX) FROM INSULATED METAL TARGETS WITH 700 keV PROTONS

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Summary: Excessive X-ray yields were observed from pure metals surrounded by insulating material, with the use of low-energy protons. These yields are affected by the proton current. An interpretation of the mechanism is given.

Key words: Charge-induced X-rays, Insulated metal targets, Protons.

Original studies[1-3] reporting enhanced X-ray yields from insulators, under bombardment with charged particles, was extended to metal targets surrounded by insulating material. Specially prepared metal discs of selected transition elements were embedded in MACOR (dielectric constant 5.9) target holders and irradiated with 700 keV protons. Improved X-ray yields of up to more than 300% were observed. It was found that the enhanced yields were affected by the incident beam current due to a "leakage" effect. The recorded time-dependent spectra showed similar charge build-up and discharge patterns as those originally obtained for insulators[2]. An interpretation of this method of improved X-ray production has been proposed and involves the build-up of charge on the insulated metal sample followed by the acceleration of energetic electrons (in the sample vicinity) towards the sample, with subsequent bombardment, up to and at the point of discharge.

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MULTITRACER --- A TRACER TECHNIQUE DEVELOPED BY USING RIKEN RING CYCLOTRON

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Summary: A versatile radioactive 'multitracer' technique has been developed at RARF. It enables efficient and simultaneous tracing of a number of elements under identical experimental conditions. Its principle and features are presented with examples of recent application.

Key words: multitracer, heavy-ion accelerator, separation, environment, bio-trace elements.

RIKEN Accelerator Research Facility (RARF) has a heavy-ion accelerator complex consisting of a ring cyclotron (RRC) and a couple of injectors: a linac and an AVF cyclotron (Fig. 1). Innovative studies are pursued in the fields of nuclear physics, solid state physics, atomic physics, nuclear chemistry, and radiation biology. In nuclear chemistry, emphasis has been put on development of the multitracer technique[1] and the in-beam Mössbauer spectroscopy in recent years.

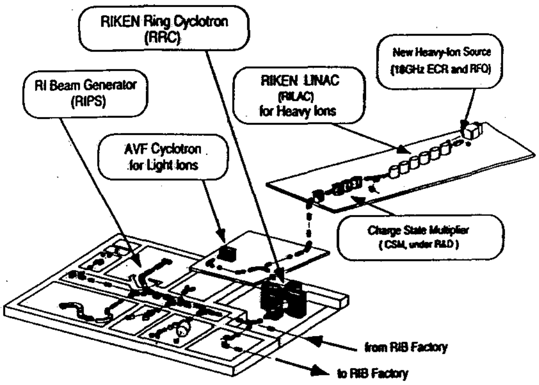

Fig. 1 The present state of RIKEN Accelerator Research Facility (RARF): A ring cyclotron and two injectors (a linac and an AVF cyclotron).

The RRC can accelerate heavy ions up to 135 MeV/nucleon. Such energetic heavy ions produce a number of radioactive nuclides at once in a target by nuclear fragmentation. We have developed the multitracer technique, in which the radioactive nuclides are used together to trace simultaneously the behavior of many elements. The technique is applied to a variety of chemical and biological investigations.

The multitracer technique enables us not only to acquire efficiently data for various elements, but also to determine the characteristic behavior of different elements under identical experimental conditions. The principle is to irradiate a metal target by high-energy heavy ions, to dissolve it in acid, and to remove chemically the target material yielding a solution of a number of radioactive isotopes, namely a multitracer solution. The solutions can also be salt-free, if an appropriate separation procedure is employed.

A target stack of gold, silver, and other metals are irradiated with a 135 MeV/nucleon beam of C, N, or O ions accelerated by RIKEN Ring Cyclotron. The irradiated gold foil is dissolved in aqua regia. The solution is evaporated to dryness under a reduced pressure in a closed vessel with a trap. The residue is dissolved in 3 mol dm^{-3} HCl and Au(III) is removed from the solution by extraction with ethyl acetate. Radioactive nuclides of Br, Sn, I, Re, and Os are recovered from the solution in the trap. The irradiated Ag foil is dissolved in (1:1) HNO_3 and silver is simply precipitated as AgCl with conc. HCl and filtered. The multitracers thus prepared are subjected to various chemical and biological experiments. The γ-rays of radioactive nuclides in the samples are measured by means of pure Ge detectors. Assignment of each peak of the γ-ray spectra to different nuclides is made on the basis of its energy and half-life.

A wide spectrum of radioactive nuclides are produced by irradiation of a target with high-energy heavy ions depending on the kind and energy of the heavy ions and the target material. Both neutron-deficient and neutron-rich nuclides are produced, the former being dominant, with cross sections for isobars amounting roughly to 10 to 100 mb. These nuclides are mainly the products of target fragmentation, covering elements with atomic numbers up to that of the target. Radioactive nuclides with an atomic number larger by one than that of the target are also produced as a result of proton transfer. Mass yield distribution, recoil properties, and angular distributions of the reaction products have been extensively studied for various combinations of heavy-ion projectiles and target elements. However, there had been no attempt before ours to utilize these mixtures of nuclides for chemical and other purposes.

The kind of elements traceable by the nuclides in the multitracer solution obtained from a target depends on time after the irradiation, radionuclides of different half-lives being produced. Because of the limited resolution of the detectors available at present, overlap of γ-ray peaks is inevitable in case of targets of heavy metals such as gold. Overlapped peaks can be resolved into two or more components by analyzing the decay curve. But, reliability of the data obtained by such analysis is not necessarily high and many of them must be discarded. Therefore, radiochemical procedures for separating the multitracer obtained from a gold target into two or three groups of elements (group tracers) have been developed. The elements traced so far by the multitracer technique are shown in Fig. 2.

The multitracer technique has been applied to various researches in the fields of basic chemistry, environmental chemistry, and chemistry of trace elements in plants and animals in cooperation with many research groups of universities, colleges and research institutes. Among the subjects studied are:

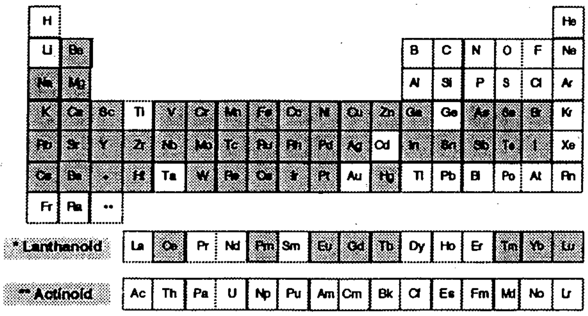

Fig. 2 Elements traced so far by the multitracer technique (shadowed ones).

(1) Adsorption of metal ions on α-Fe_2O_3,
(2) The influence of humate formation on the adsorption of various ions on kaolinite and silica gel,
(3) Sorption of platinum group metals onto agricultural soils,
(4) Removal mechanism of metal elements from sea water,
(5) Transport of metal ions in rice and soybean plants,
(6) Behavior of various elements in atmosphere-plant system,
(7) Distribution and excretion of various elements in normal rats,
(8) Bone accumulation of vanadium,
(9) Trace elements in zinc deficient rats,
(10) Biochemical properties of platinum-group elements,
(11) Comparative uptake behavior of trace elements in normal, Al-overloaded and Cd-overloaded mice,
(12) Metabolic behavior of rare earth elements in non-insulin dependent diabetes Mellitus mice,
(13) Biodistribution of trace elements in Wistar and LEC rats,

(14) Labeling of immunoglobulin G with a multitracer,
which are compiled in ref. 2 along with reports on preparation of multitracers and those on mechanisms of heavy-ion reactions. Examples of more recent application will be presented in the lecture.

In conclusion, the results described above demonstrate the versatility of the multitracer technique in studying the behavior of a number of elements in various systems. The multitracer technique has opened a new way of application of heavy-ion physics to chemistry, biology and medicine.

Now, RARF undertakes construction of an RI-Beam Factory (RIBF) capable of providing the world's most intense RI beams at energies of several hundreds MeV/nucleon over the whole range of atomic masses. This Factory is expected to realize a new era of not only nuclear physics, but also nuclear and radiochemistry, including development of new tracer techniques.

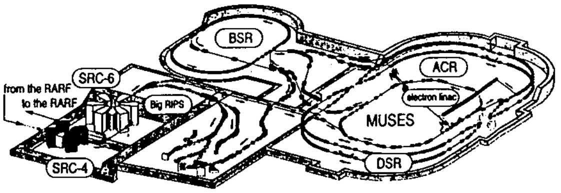

Fig. 3 RIKEN RI-Beam Factory (RIBF, a new plan just started).

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