The Abdus Salam International Centre for Theoretical Physics

2484-19

ICTP-IAEA Joint Workshop on Nuclear Data for Science and Technology: Medical Applications

30 September - 4 October, 2013

α, β+, γ and Auger-electron Decay Data in Nuclear Medicine − Experimental Determination, Status and Deficiencies Part I

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Guildford
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α, β^+, γ and Auger-electron Decay Data in Nuclear Medicine − Experimental Determination, Status and Deficiencies
Part I

Alan Nichols

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Manipal University, Madhav Nagar, Karnataka, India

30 September 2013

Workshop on Nuclear Data for Science and Technology: Medical Applications
ICTP, Trieste, Italy, 30 September – 4 October 2013

Alan Nichols ?

Alan Nichols

1974-1977	**UKAEA, AERE Harwell, UK**
	variable energy cyclotron
	nuclear decay data
1977-1993	**UKAEA, AEE Winfrith, UK**
	aerosols, radioactive waste management
	nuclear data
1993-2001	**UKAEA/AEA Technology, Harwell and Culham, UK**
	various management functions
2001-2009	**IAEA, Vienna, Austria**
	nuclear data (Nuclear Data Section)
2009	**University of Surrey, UK; Manipal University, India**
to date	**sole trader, UK**
	nuclear data assessments, decay data evaluations

Alan Nichols

evaluation of nuclear data ….

and decay scheme data in particular

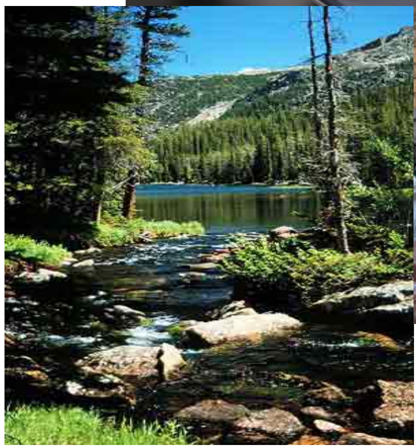
IAEA Mandate

"Atoms for Peace"

To contribute to sustainable development in Member States by the use of nuclear sciences and their applications in food and agriculture, human health, industry, water resource management and environmental monitoring and protection.

Nuclear Sciences and Applications:
Serving Basic Human Needs



Alan Nichols

ICTP, Trieste, Italy, 30 September – 4 October 2013

Context:

- 850 million suffer from chronic malnutrition, six million child deaths per year
- Six million die annually of cancer, 10 million new cases each year
- 1.2 billion lack access to safe, secure water supplies
- Bacterial, parasitic and viral infections kill millions per year e.g. maleria kills 2 million annually
- Climate change, with negative impacts on ecosystems, water resources, agriculture and health

Nuclear techniques can address specific issues:

- ✓ Provide better health care
- ✓ Improve agricultural productivity and food security
- ✓ Improve management of scarce water resources

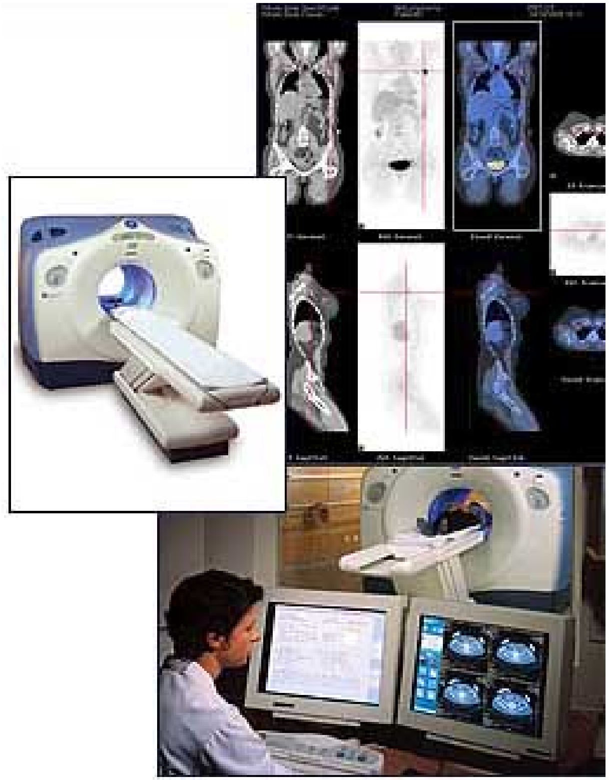
- ✓ Improve understanding and management of the environment

Atoms for Health: Disease Prevention and Control
Division of Human Health

- **Nutrition**
- **Nuclear Medicine**
- **Radiobiology and Radiotherapy**
- **Dosimetry and Medical Physics**
- **Fighting Global Cancer**

Nuclear Medicine

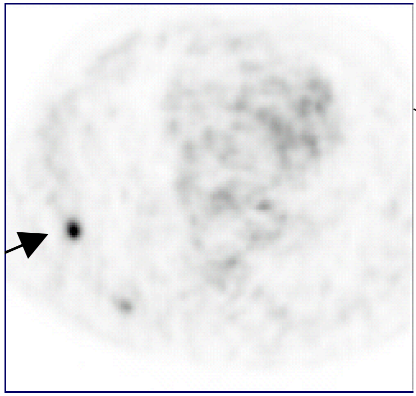
- Nuclear imaging techniques enable
  accurate and detailed diagnoses
- Optimized treatment of illnesses
  such as cancer and cardio-
  vascular disease
- Objectives
  - improve integration of nuclear technology and planning in treatment of disease
  - enhance human resources capacity (e.g. physicians, physicists, radiopharmacists)



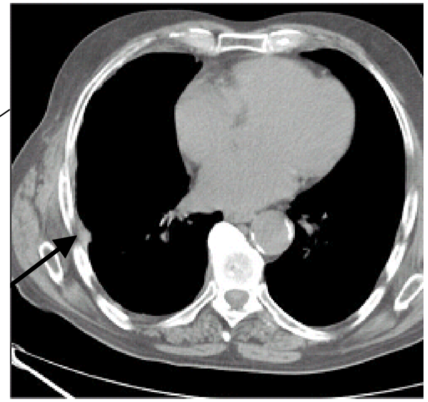
Combined PET-CT machine

Nuclear Medicine

Improved cancer diagnosis using ‘fusion’ of PET (left) and CT (right) images

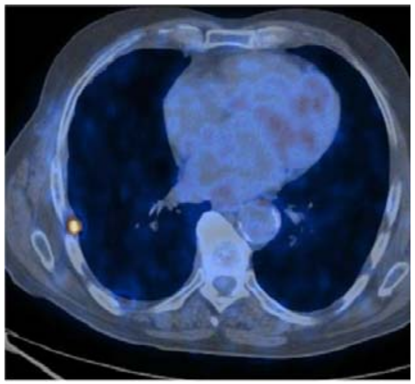


Positron Emission Tomography



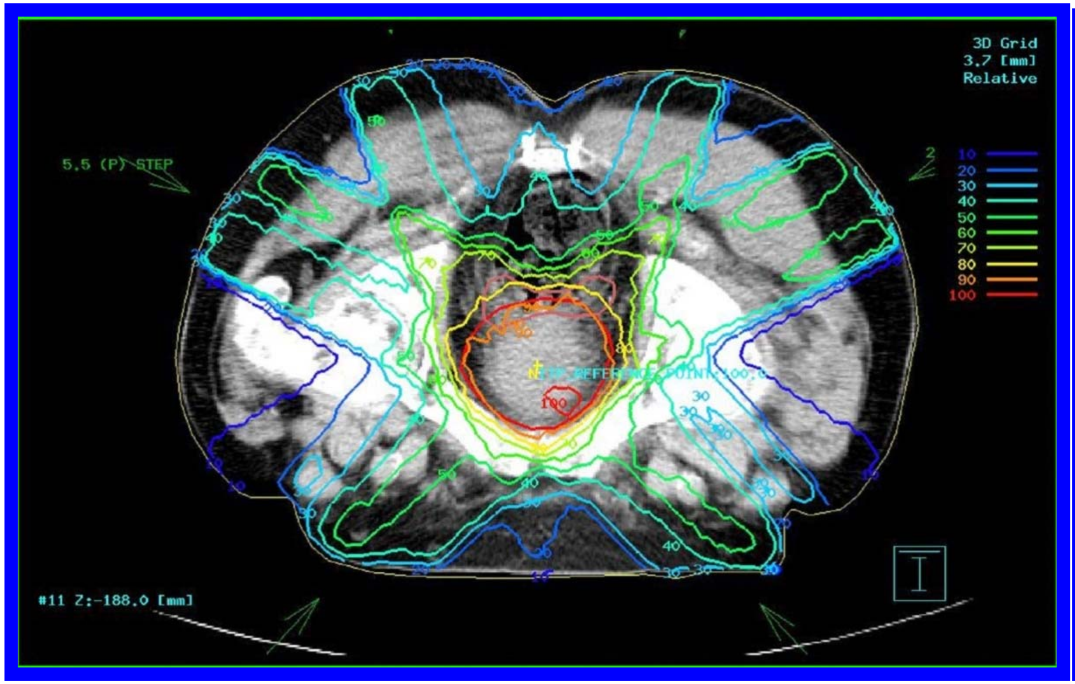
Computerised Tomography

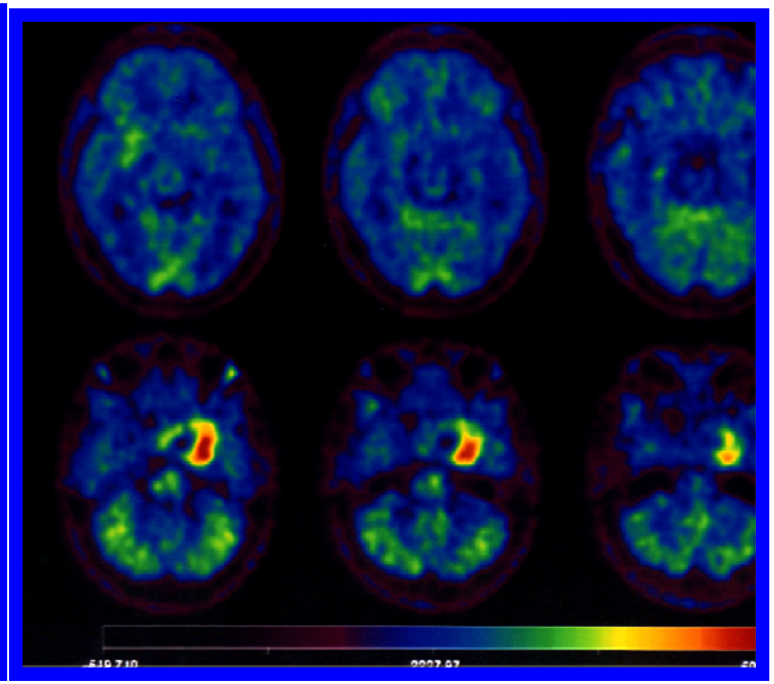
combined image



Nuclear Data for Medical Applications

Rationale: **Cancer therapy** **Diagnosis**



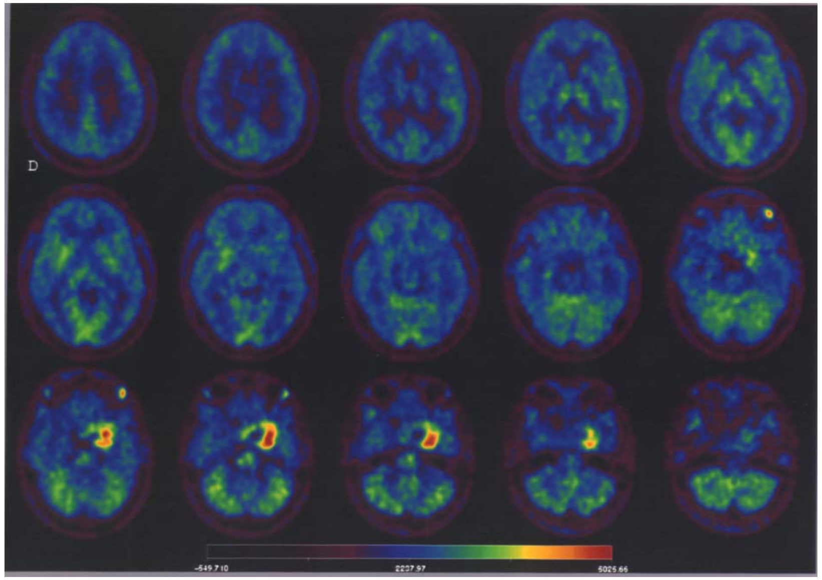


Beneficiaries: **medical physicists, radioisotope producers, researchers …**

Objectives: **improve data for medical radioisotope production and patient dose delivery calculations in radiotherapy**

ATOMIC AND NUCLEAR DATA

SUPPORT FOR MEDICAL APPLICATIONS



- Production of radioisotopes for medical diagnosis and therapy
- Radiotherapy: photons, electrons, protons and heavy ions
- Brachytherapy combines radiotherapy with monoclonal antibodies
- Dosimetry and radiation safety
- Radiation transport data

Radiation Biology and Radiotherapy

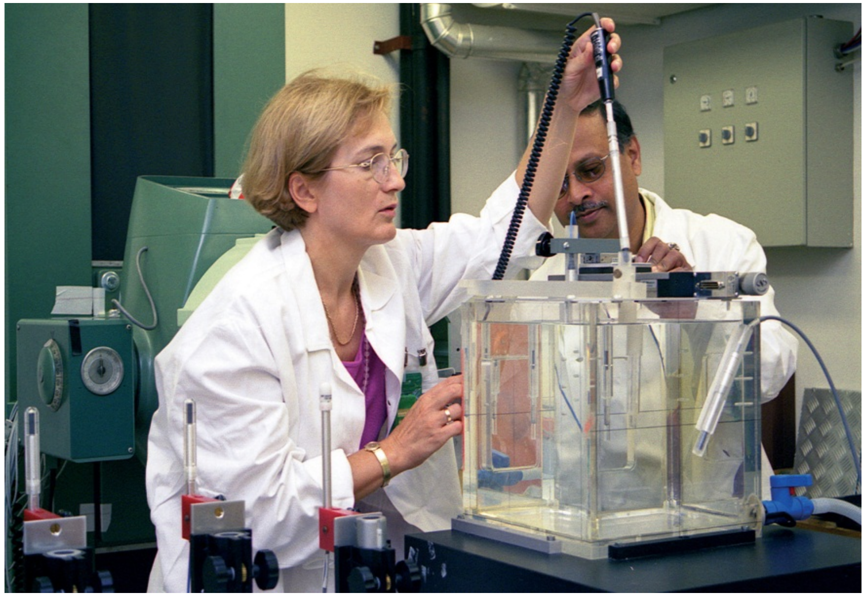
External beam radiotherapy

Improve the availability and safe use of cancer management techniques in Member States by:

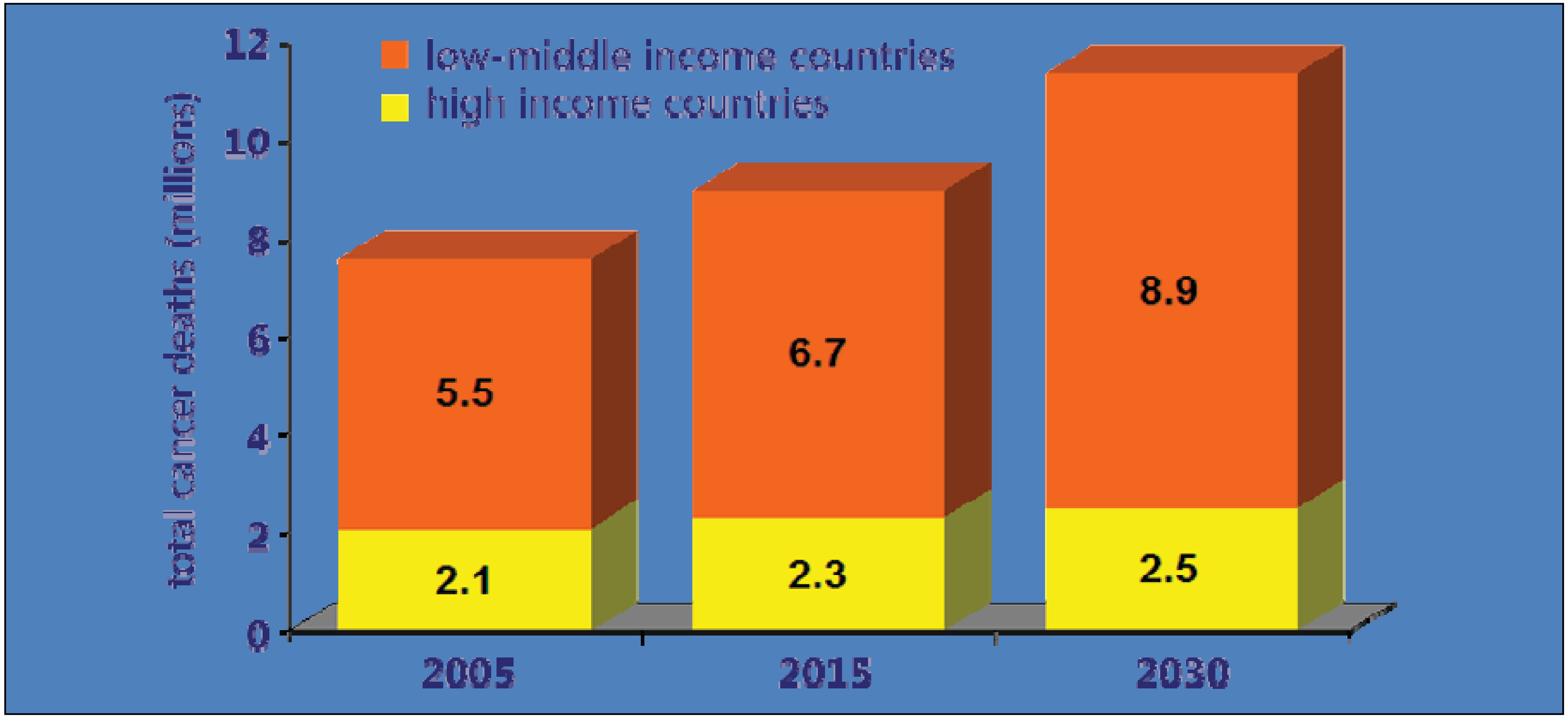
- helping to establish new treatment centres
- organize teaching and training courses
- devise more effective use of radiation in cancer treatment

Medical Physics and Dosimetry

- Quality and safety assurance in medical uses of radiation
- Dosimetry calibration services to ensure accuracy and comparability of results
- Establishment of international standards to harmonize medical uses of radiation



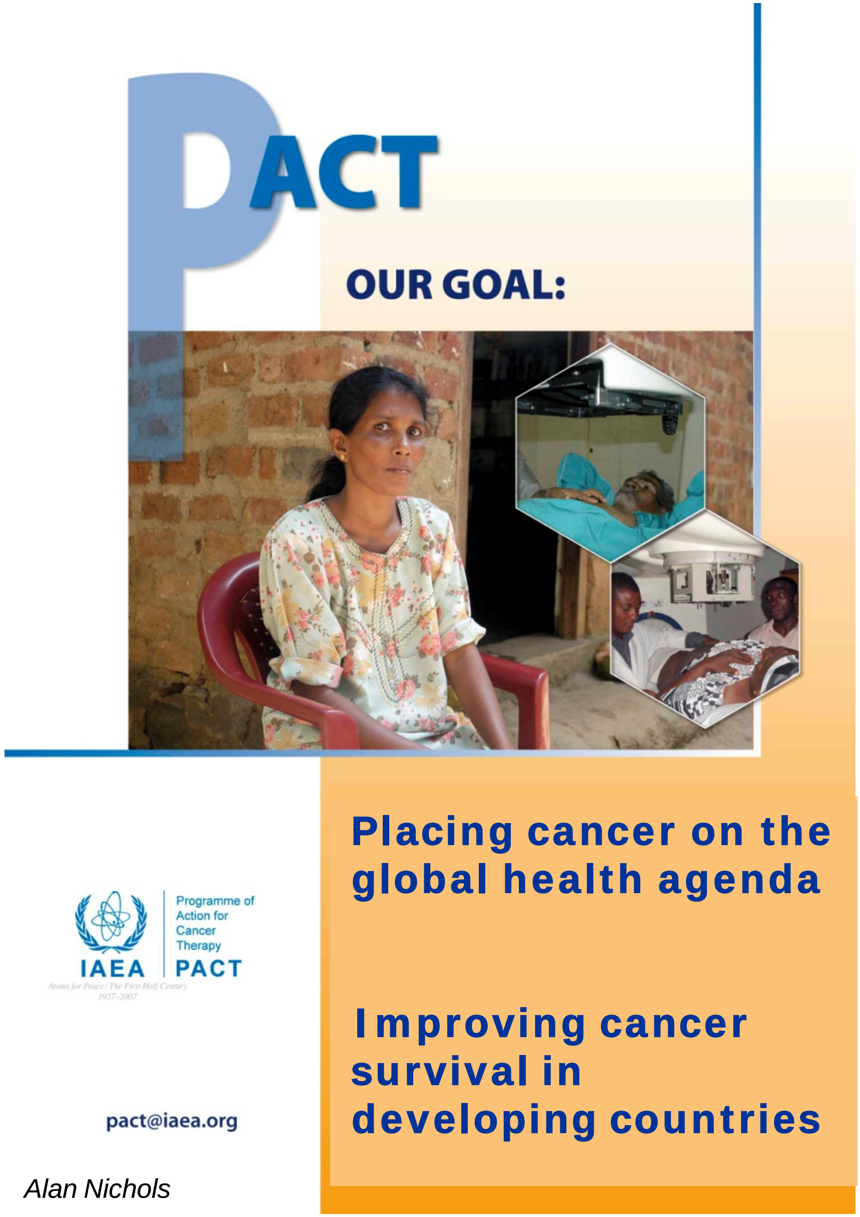
Fighting Global Cancer



- approximately 260 million new cancer cases expected in 20 years
- nearly 150 million will occur in developing countries
- cancer: no. 1 killer

Graph: WHO (2008)

Programme of Action for Cancer Therapy (PACT)



- **PACT was created to help expand radiotherapy capacity in developing countries, and improve the quality of and access to cancer care services:**
  - integrate IAEA radiotherapy assistance into **public health and cancer control systems**
  - establish a well coordinated IAEA cancer programme with **harmonised strategy**
  - advocate a country level, **bottom-up approach**

PACT: strategy

1. **Capitalize on IAEA expertise in radiotherapy**

2. **Build and maintain partnerships with WHO, development funders, NGOs, and private sector**

3. **Mobilize resources for specific projects**

Implementation Mechanisms: Coordinated Research

Applied research: **research to evaluate technology or concepts, and determine whether these can be usefully "applied", with or without further refinement, to achieve development objectives**

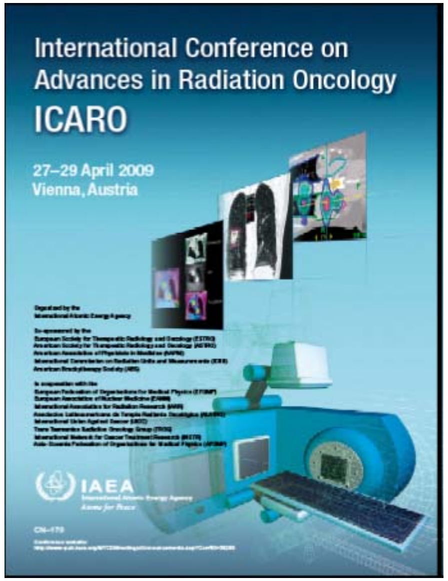
Adaptive research: **research to adapt already existing and well-tested knowledge or technology for adoption by beneficiaries ("local tailoring").**

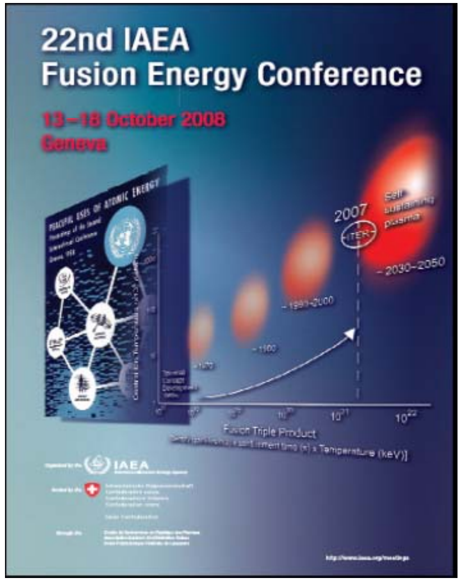
Coordinated Research Projects (CRP)

Promote research, development and transfer of nuclear technology

- **Member State scientists choose to share research tasks to achieve a common goal**
- **Project formulation by participating scientists and the IAEA to ensure relevance to IAEA programmes**
- **Research conducted by Member State scientists with IAEA acting as coordinator**
- **Meetings held every 18 months to discuss work plan and progress**
- **Report and publication of achievements upon completion**

Implementation Mechanisms: Meetings and Publications



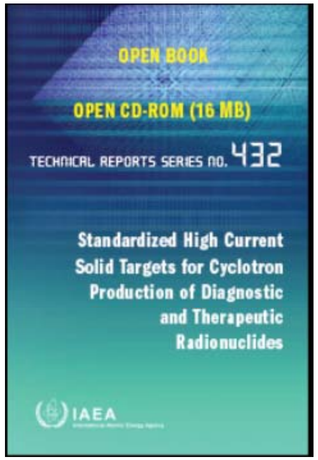


Meetings:

- symposia
- workshops
- advisory groups
- seminars, etc.

Publications:

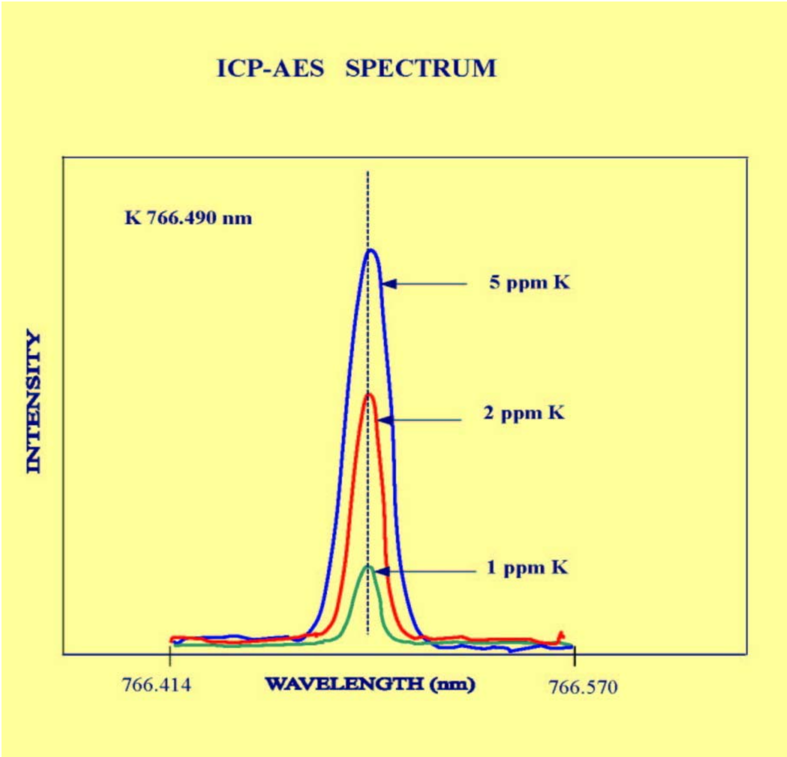
- proceedings
- technical documents
- manuals
- Standard Operating Procedures



Implementation Mechanisms: Database Services

IAEA plays a key international role as a repository and provider of scientific nuclear data and knowledge

- ✓ Fission reactors
- ✓ Fusion
- ✓ **Medicine**
- ✓ Water resources
- ✓ Atmospheric and marine data



Atomic and Nuclear Data

1. Reaction data

→ neutron, charged-particle and photon cross sections

→ interactions with target nuclei, atoms and molecules as a function of energy of projectile

ALADDIN: atomic and molecular data

EXFOR: neutron and charged-particle data

Atomic and Nuclear Data

2. Nuclear structure and decay data

→ nuclear energy levels, half-lives and radioactive decay (α, β, γ)

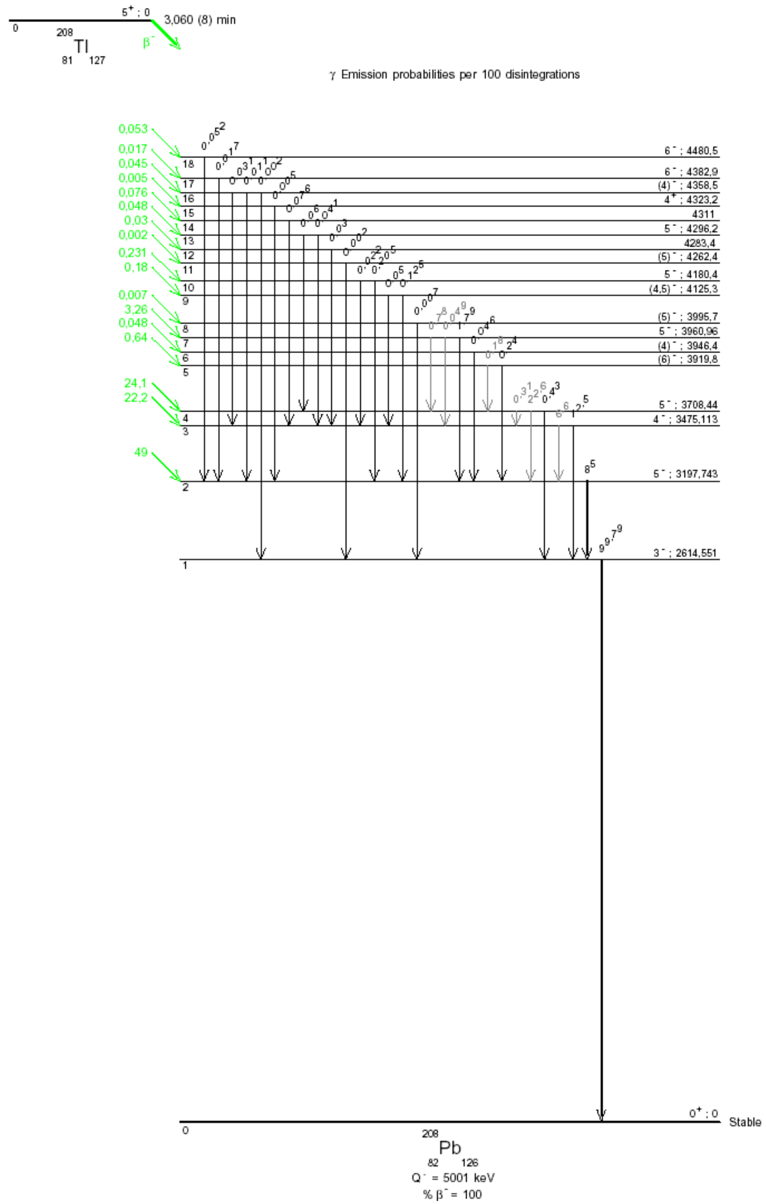
ENSDF

MIRD

DDEP

3. Atomic decay data

→ X-rays, Auger electrons


5+ ; 0
3,060 (8) min
208 Tl 81 127
β-
γ Emission probabilities per 100 disintegrations
6- ; 4480,5
6- ; 4382,9
(4)- ; 4358,5
4+ ; 4323,2
4311
5- ; 4296,2
4283,4
(5)- ; 4262,4
5- ; 4180,4
(4,5)- ; 4125,3
(5)- ; 3995,7
5- ; 3960,96
(4)- ; 3946,4
(6)- ; 3919,8
5- ; 3708,44
4- ; 3475,113
5- ; 3197,743
3- ; 2614,551
0+ ; 0
Stable
208 Pb 82 126
Q- = 5001 keV
% β- = 100

Major NSDD products

ENSDF - ***www.nndc.bnl.gov/ensdf*** - **J.K. Tuli**, ***NNDC***

Contents: Evaluated nuclear structure and decay data for all known nuclei, organized into over 290 mass chains

ENSDF

Decay data

β^-
ε/β^+
α
β^-n
etc.

Reaction data

(HI,xnγ)
(p,p')
(n,γ), (n,xnγ)
Coulomb Exc.
(α,α'), (α,xnγ)
(d,p), etc.

Adopted data

best values

Levels:

E, J_{π}, $T_{1/2}$, μ, Q, configurations

Gammas:

E_{γ}, BR, M, δ, ICC

ENSDF: Major Data Sources and Derivatives

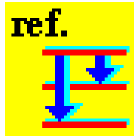
Contributing Databases:

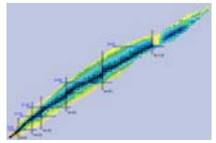
- Atomic Masses (Audi, Wang)
- Nuclear Science References (NSR)

ENSDF

MC Codes
- ❑ MCNPX
- ❑ GEANT

ORTEC & CANBERRA





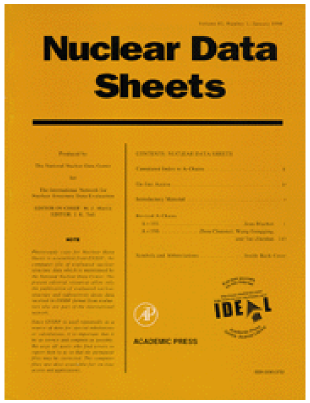
IE

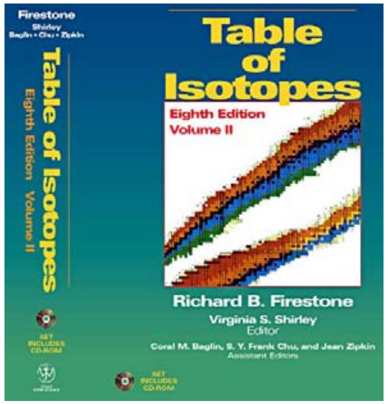
NuDat2

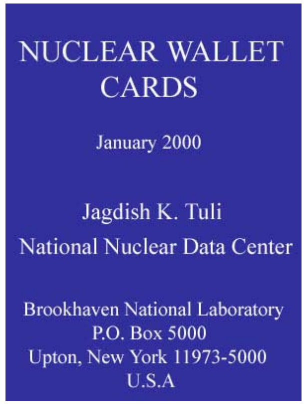
Derivative Databases:

NuBase | MIRD | RIPL | JANIS | RADWARE | …

Derivative Publications:







Isotopes (what, where and why?)

- **Stable and radioactive** isotopes play critical roles in a variety of technological applications important to humanity in a modern society:
  - ✓ Nuclear medicine:
    - diagnosis
    - radiotherapy
    - imaging
    - external radiation sources
  - ✓ Power sources (e.g. nuclear batteries)
  - ✓ Oil industry
  - ✓ Tracers
  - ✓ Many other commercial applications
  - ✓ National Security
  - ✓ Basic scientific research

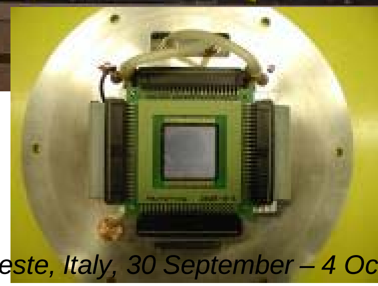
$^{99}Tc^{m}$

$^{238}PuO_2$

- Production of **radioactive** isotopes involves either **nuclear reactors** or charged-particle **accelerators**

Counting Laboratory Detectors

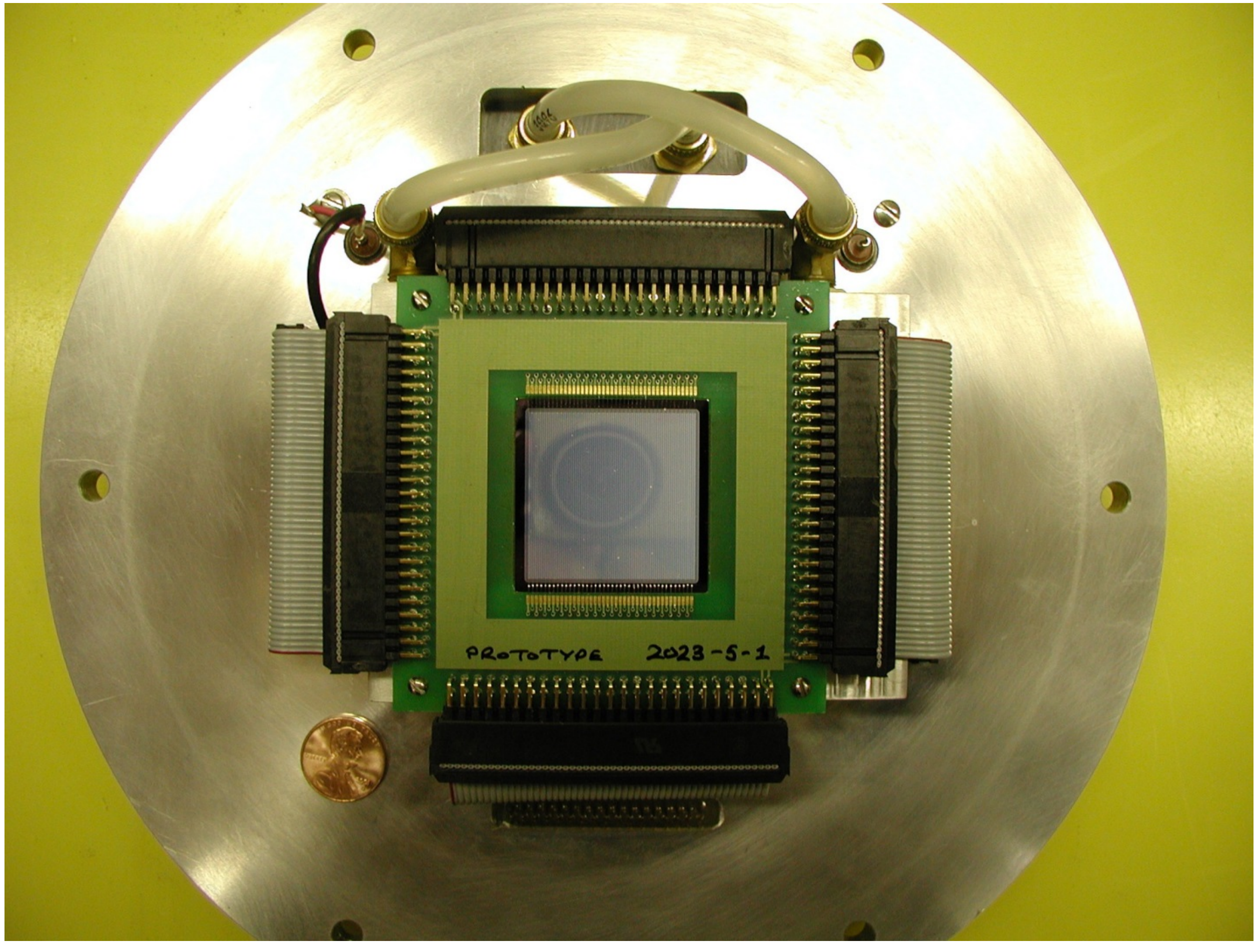
- Germanium detectors – single Ge crystals, LEPS (low-energy photon spectrometry), CLOVERS, DSSD – various size, high efficiency and energy resolution
- Si detectors (including DSSD) and PIPS (passivated implanted planar silicon) – mostly used for CE or β particles, but also for p– and α–decay studies
- Plastic scintillators, La_2Br_3, etc.
- Alpha-particle spectrometry
- Electron spectrometry



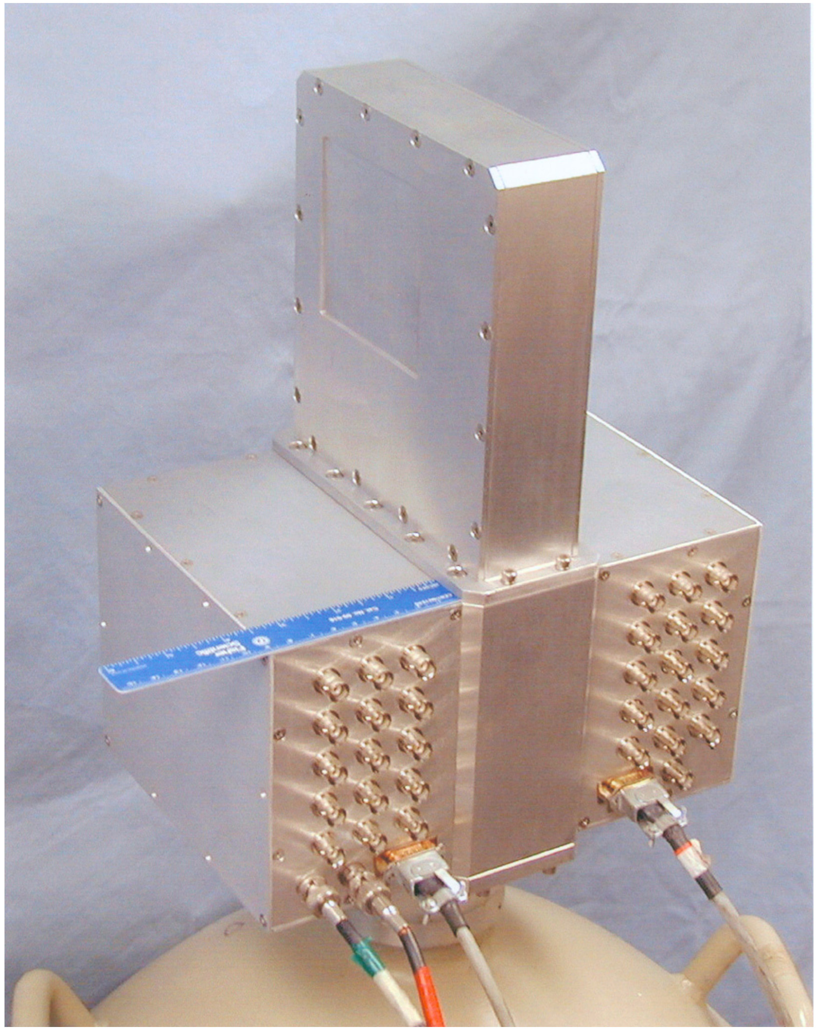
able to carry out precise measurements of relevance to medical isotope research by means of a variety of experimental techniques and detector systems

Counting Laboratory Detectors: HPGe

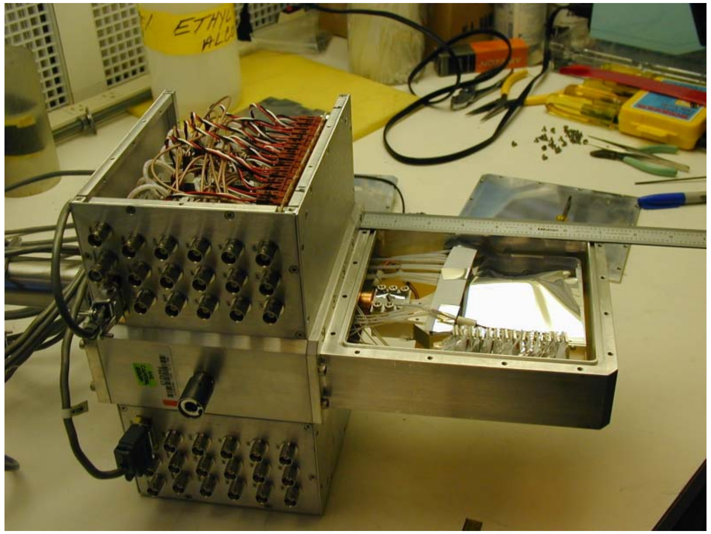
Counting Laboratory Detectors: Si-DSSD (silicon Double-Sided Strip Detector)

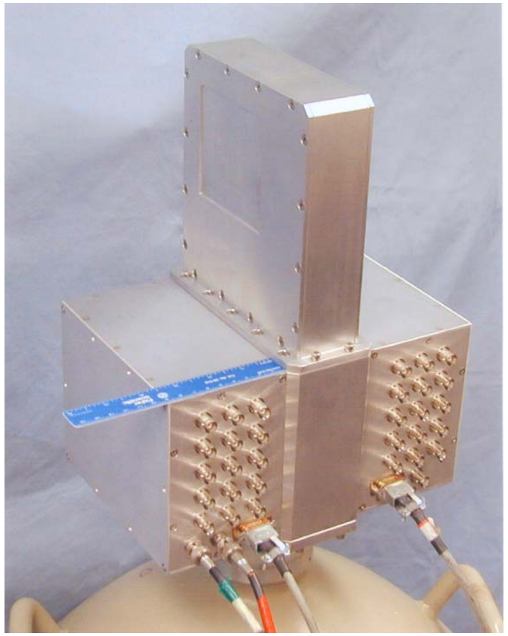


Counting Laboratory Detectors: Ge-DSSD (germanium Double-Sided Strip Detector)



HPGe Strips Detector





built by **Ortec** and **ANL**

large ~ 90 mm x 90 mm x 20 mm

resolution ~ 1.0 keV at 122 keV; ~ 2.0 keV at 1.3 MeV

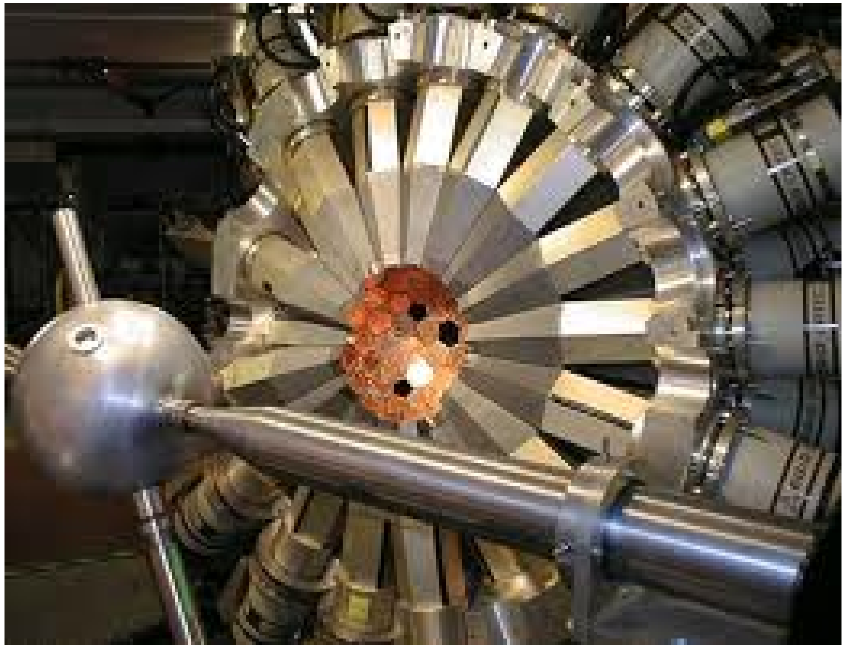
Ge strips detector

Nuclear Decay Data

cluster arrays:

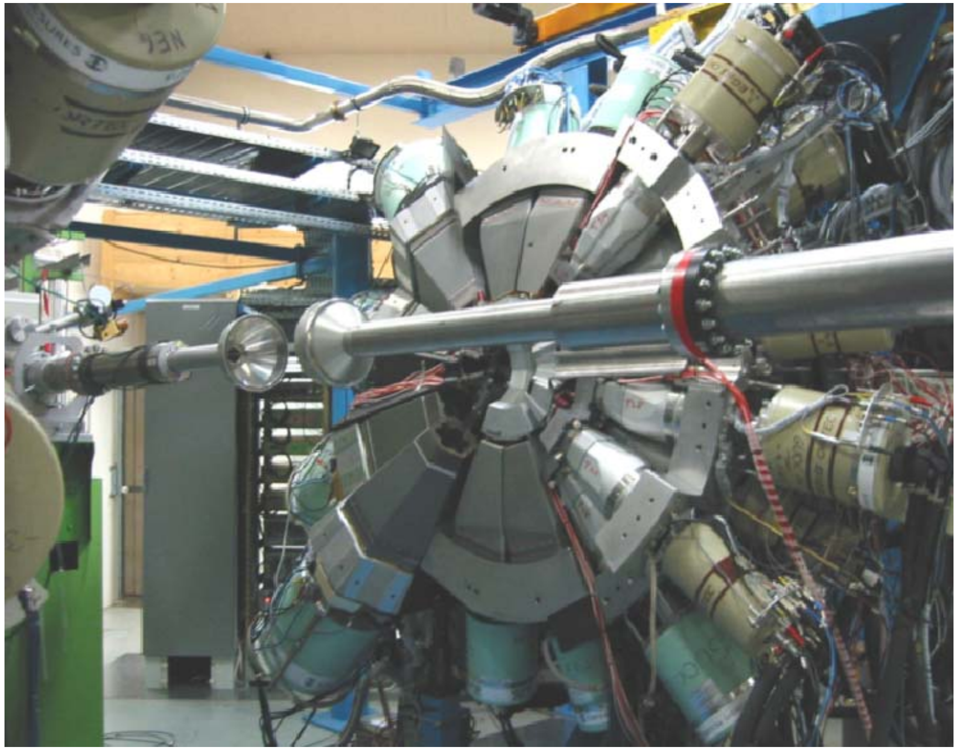
- advanced γ-ray detector technology
- resolution of complex decay schemes, astrophysics and basic nuclear physics research
- 1980s/90s – **EUROBALL**, **GAMMASPHERE**
- 2000s – **AGATA**, **GRETA** (4π), **GRETINA** (π)

High-purity Germanium Detectors: γ arrays



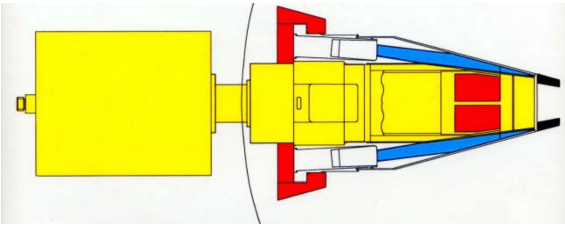
▲ Half modular layout of HPGe detectors for **Gammasphere**

Half modular layout of HPGe
▼ detectors for **Euroball**

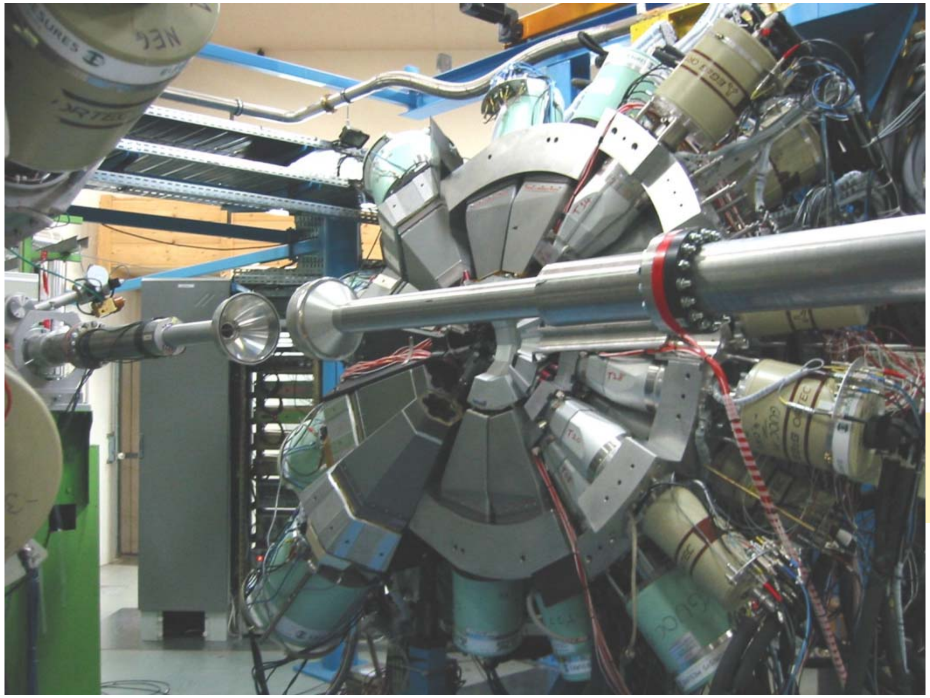


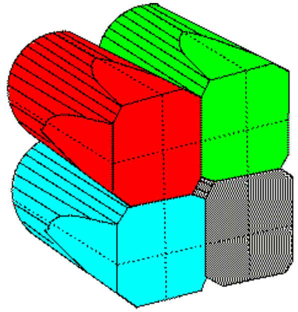
EUROBALL

European collaboration
France, Denmark, Germany, Italy, Sweden and the UK



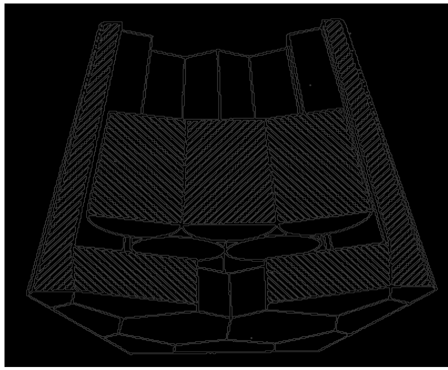
30 large single crystal
Ge detectors





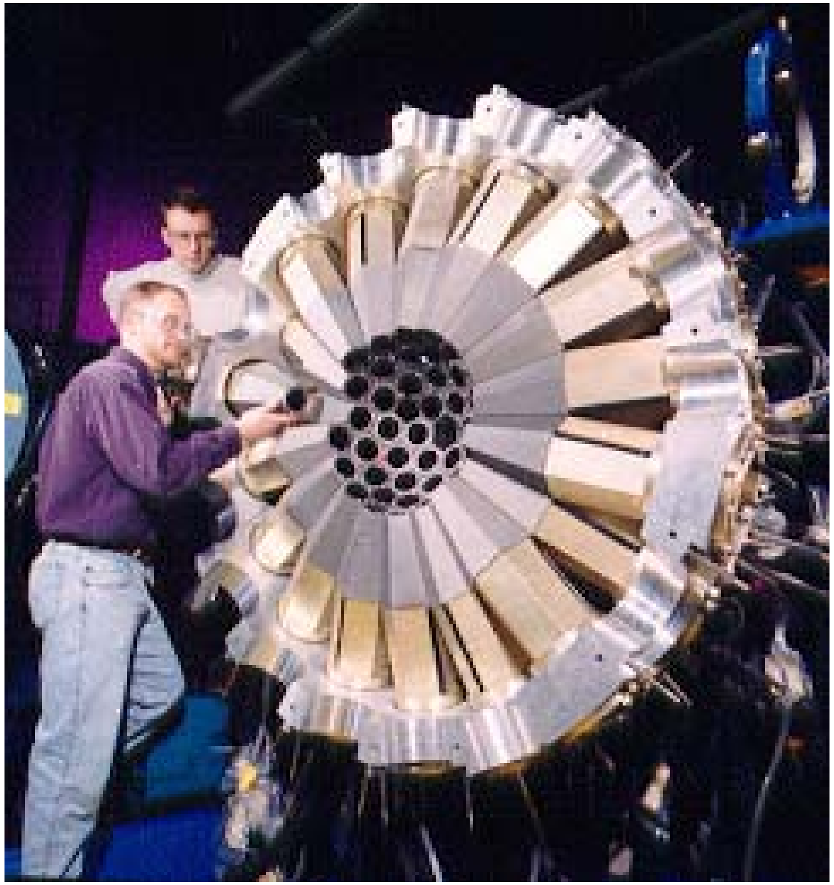
26 clover Ge detecto
4 crystals per cryostat

239 Ge crystals with
suppression shields
Total peak efficiency ~ 9.4%
Intensity limit ~ 10^{-5}



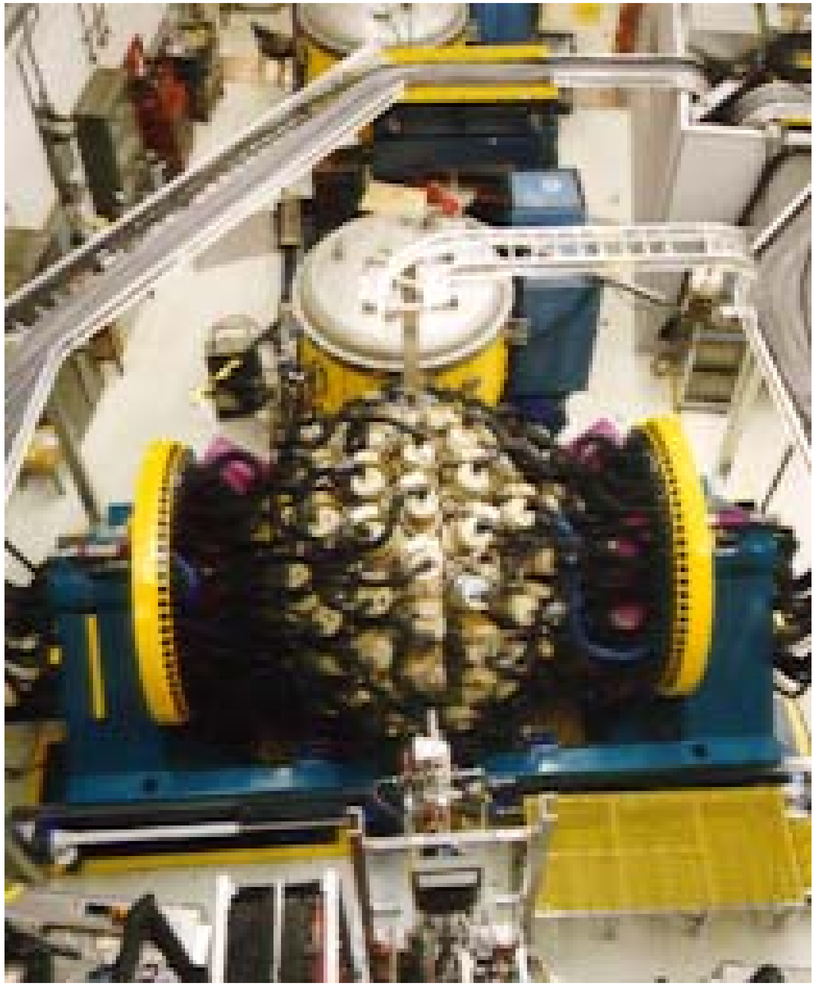
5 cluster Ge detectors
encapsulated Ge crystals per cluste

GAMMASPHERE Spectrometer



- Spectrometer with high detection sensitivity to electromagnetic radiation - high resolution, granularity and efficiency
- Consists of a spherical shell of 110 large volume HPGe detectors, each enclosed in a BGO shield
- Funded by US DoE

GAMMASPHERE Operation



- 1993 to 1997: GAMMASPHERE was constructed and located at the 88-inch cyclotron, LBNL
  - 130 experiments
  - super deformation
- 1998 to 2000: GAMMASPHERE operated at ATLAS, ANL
  - 101 experiments
  - nuclei far from stability
- March 2000 to January 2002: LBNL
- March 2002 to date: GAMMASPHERE at ANL

Nuclear Decay Data

cluster arrays:

AGATA - 4π (Europe)

GRETA - 4π (USA)

GRETINA - π (USA)

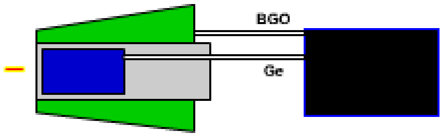
γ-ray energy tracking

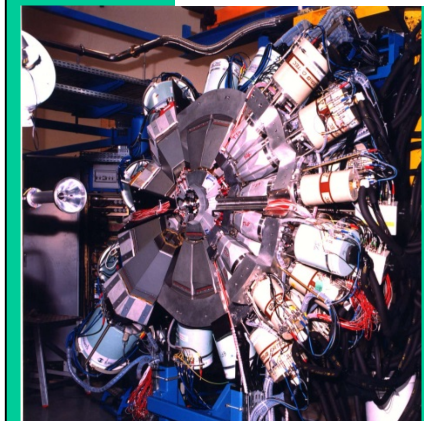
electrically-segmented Ge crystals

locate position and energy points in detector segments

HPGe strips detector

large gamma arrays based on Compton-suppressed spectrometers



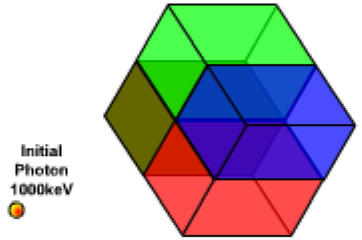


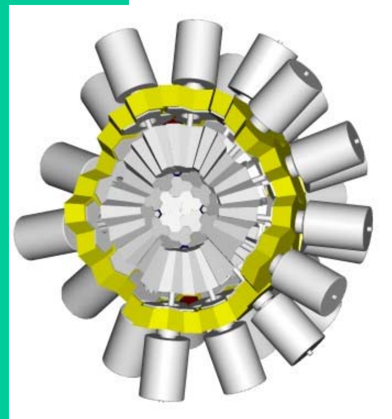
EUROBALL

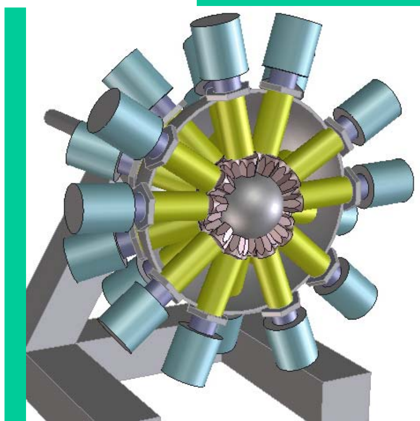
GAMMASPHERE

ε ~ 5 — 10 %

tracking arrays based on position-sensitive Ge detectors







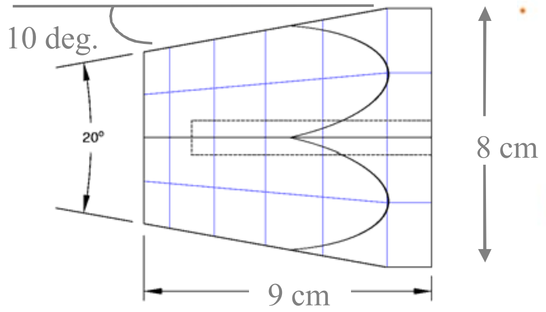
AGATA

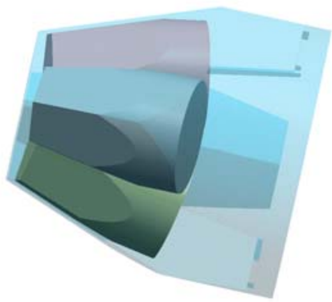
GRETA

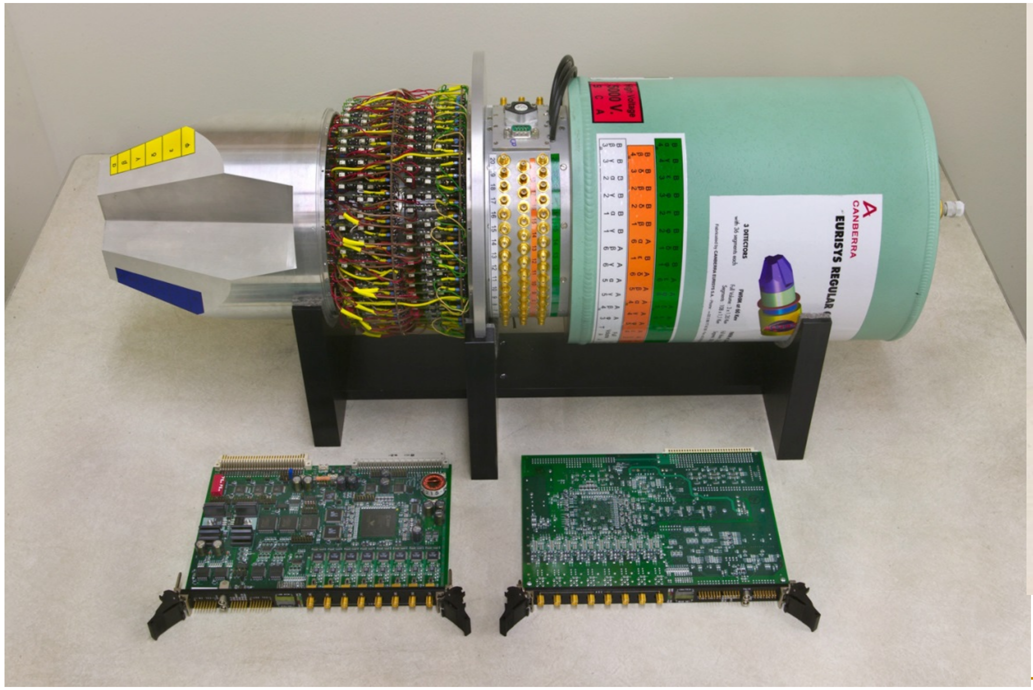
ε ~ 40 — 60 %

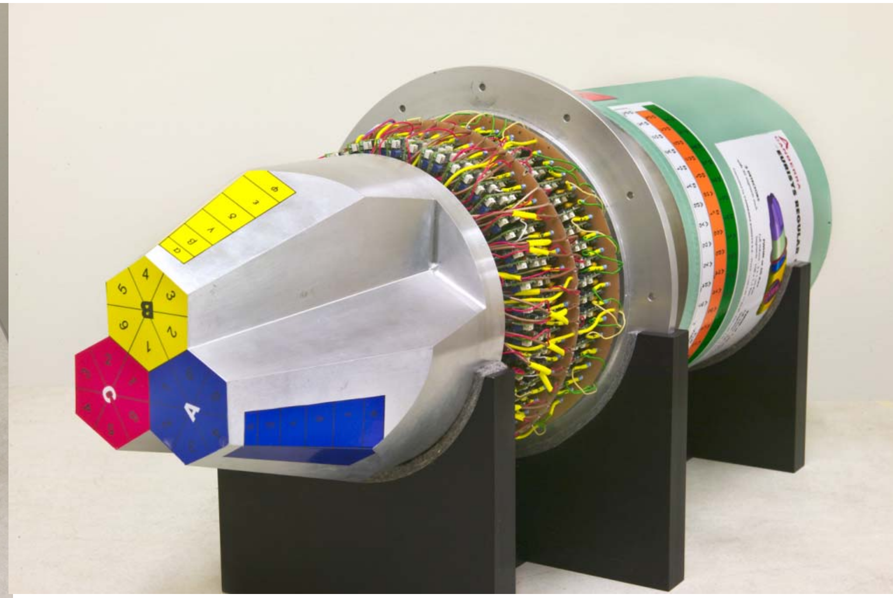
GRETINA Detectors

Tapered hexagon shape
Highly segmented, 6 x 6 = 36
Close packing of 3 crystals
111 channels of signal







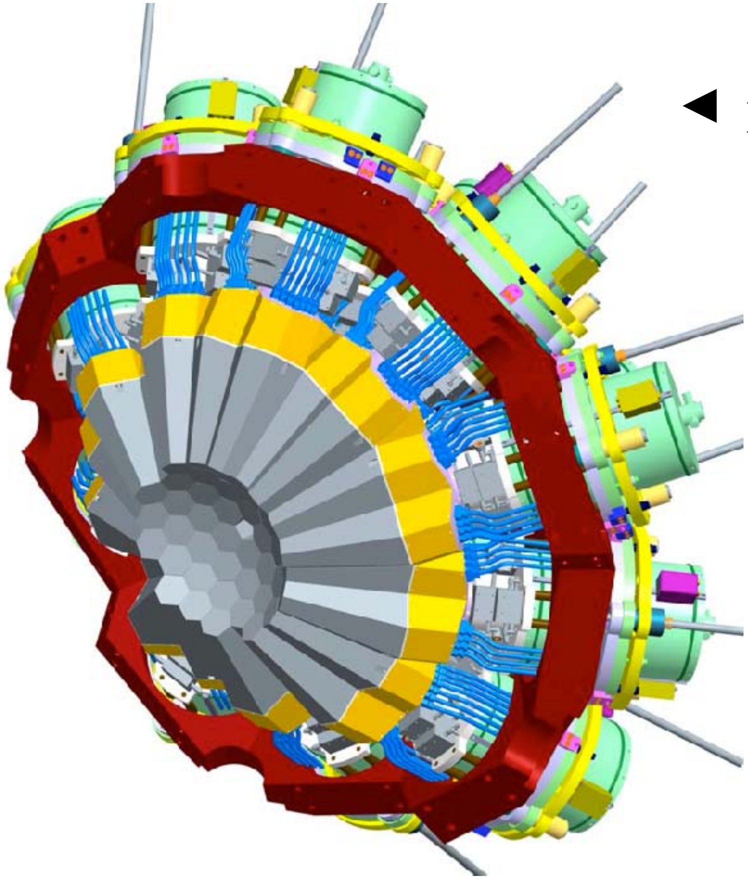


Nuclear Decay Data

cluster arrays:

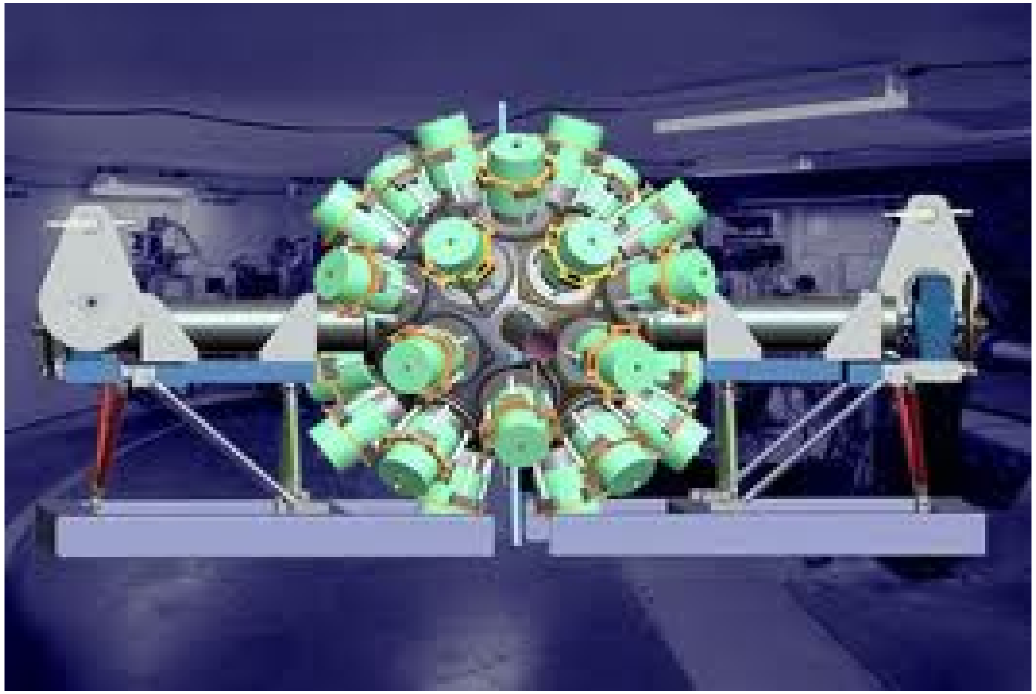
- demonstrated feasibility of γ-ray energy tracking
- represent new opportunities in nuclear medicine and basic nuclear physics research
- major response to analytical demands of the future that will arise from RIB (rare isotope beams)

21st century γ arrays: AGATA and GRETA

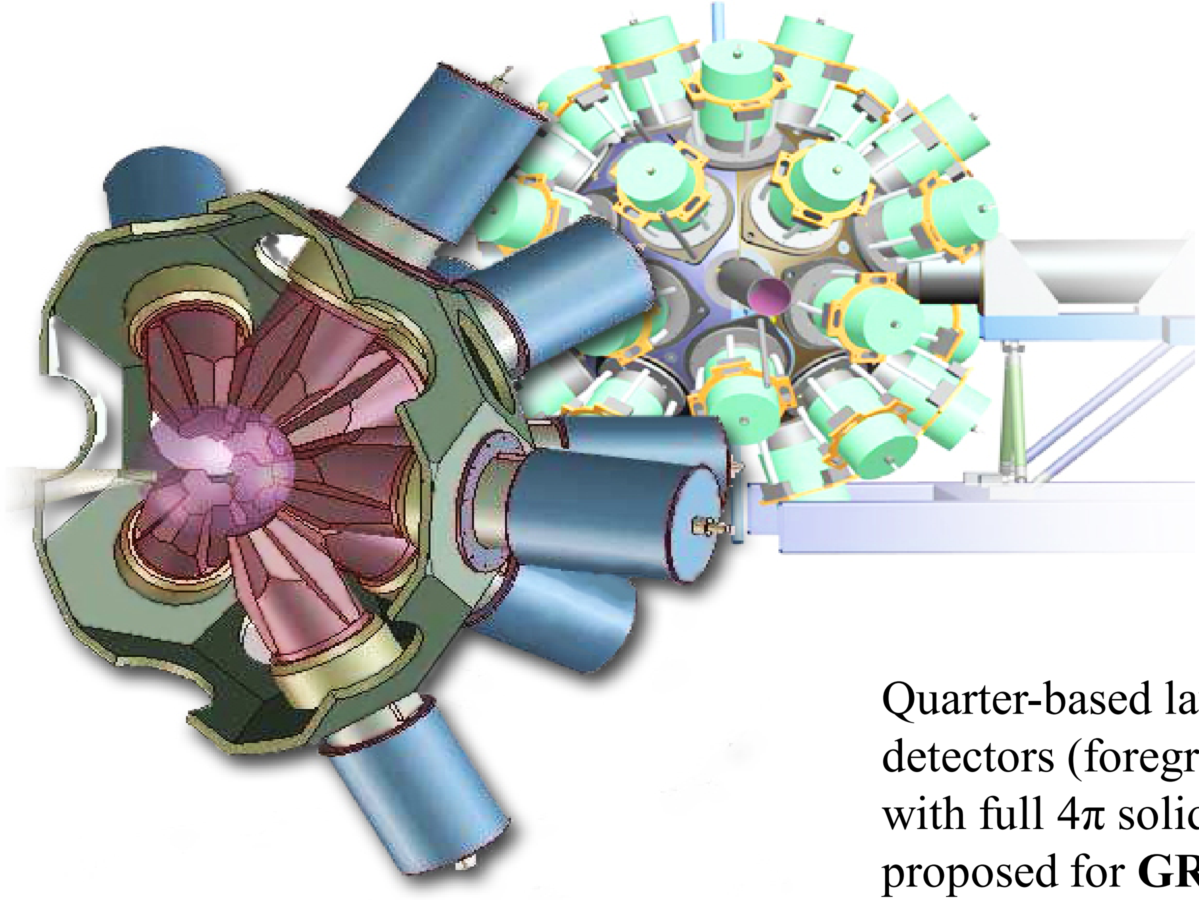


◄ partial cross-sectional arrangement for AGATA

full 4π solid-angle arrangement for GRETA ▼



21st century γ arrays: GRETINA → GRETA



Quarter-based layout of **GRETINA** detectors (foreground) compared with full 4π solid-angle system proposed for **GRETA** (background)

Nuclear Data Needs

- cross sections for various production reactions – neutrons, charged particles, photons

- nuclear decay data:
  - ✓ determination of cross sections
  - ✓ important for specific medical applications, e.g., imaging, diagnosis, therapeutic treatment, etc.

- atomic decay data associated with atomic radiations produced in radioactive decay: Auger, Coster-Kronig, super-Coster-Kronig, and other shake-off electrons

Nuclear Decay Data

- **Q values -** G. Audi, *et al*., Chin. Phys. C36 (2012) 1287-2014

- **Lifetime (**evaluated) – in good shape for most cases (exceptions, for example ^{186m}Re), but no consistent evaluation methodology

- **Emission/transition energies and probabilities (γ, β^-, β^+, α, EC, CE, Auger electrons) and relevant spectra**

  ✓ know decay scheme - E_x, J^π, multipolarity (parent-daughter)

  ✓evaluated data: energies and emission probabilities, and δ (for γ transitions) - usually several measurements, except for **g.s. to g.s. β decay** – treatment of discrepant data

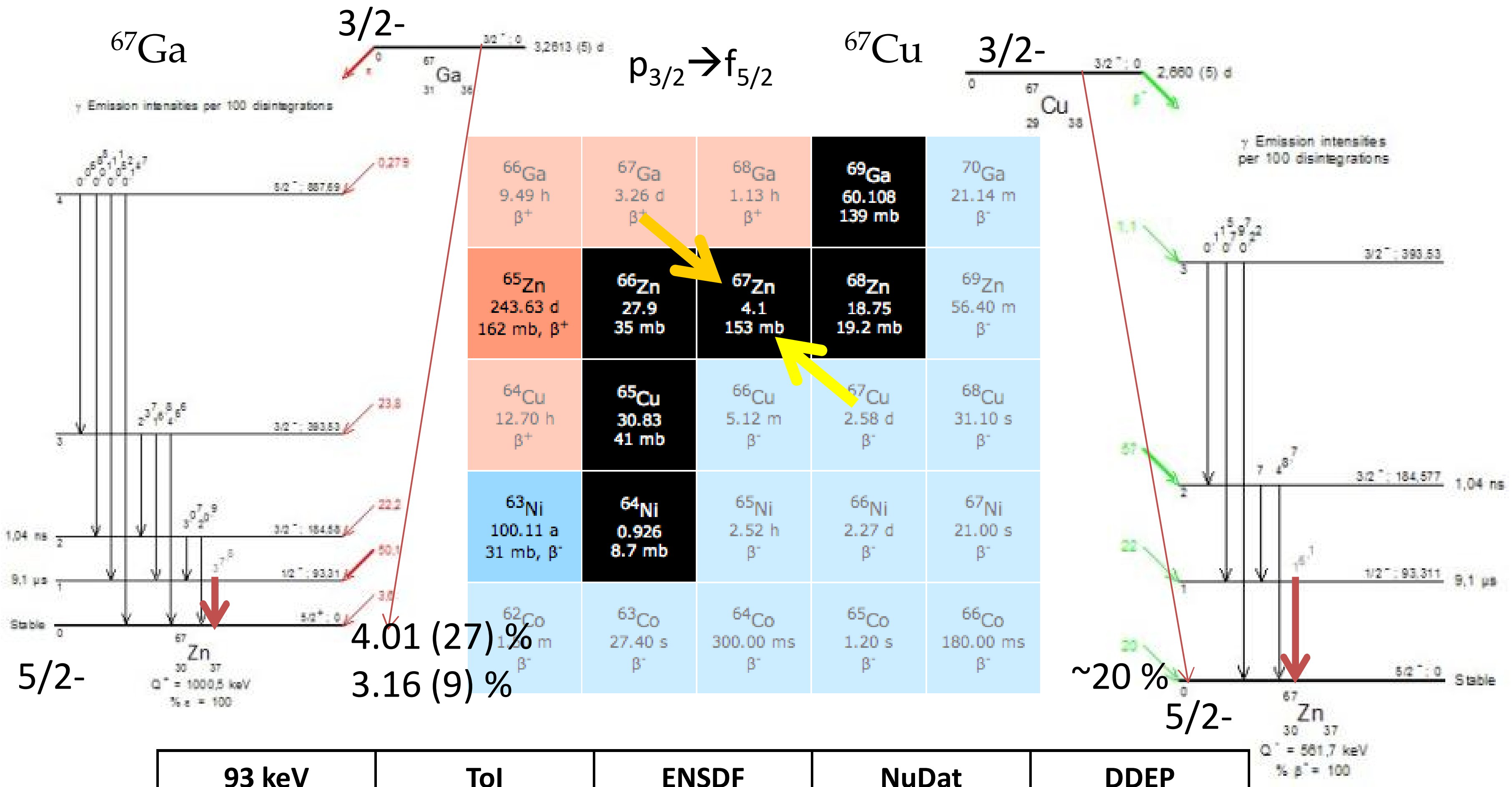
  ✓ CE data – usually from BrIcc, ….. but when multipolarities are not known, or E0?

  ✓derived data: atomic radiation, EC/β^+ ratios, β^- decay energies and emission probabilities

How good are nuclear and atomic decay data?

- answer is ambiguous
  - ✓ what are the requirements for cross-section measurements – 1%, 5%, 10% or more? … realistically perhaps 5 to 10%?
  - ✓ what are the requirements for a specific medical application – imaging, diagnosis, treatment? … perhaps better than 5%?
- status (and quality) of your favorite database – ToI (outdated, 1999 Edition based mostly on ENSDF); NuDat and MIRD – (originate from ENSDF – date of update is not given, so you cannot track origins, and sometimes conflicting information)
  → best bets are ENSDF and DDEP – both have pros & cons
- many evaluations do not provide information on problems with the data and what would be useful to measure in order to improve the reliability of the recommended decay data set (perhaps with a few exceptions – depends on evaluator's experience and knowledge)

^{67}Cu and ^{67}Ga



93 keV	Tol	ENSDF	NuDat	DDEP
^{67}Ga	39.2 (10)	38.81 (3) (0.9 (9)%)	38.81 (3)	38.1 (7) (3.6 (20)%)
^{67}Cu	16.1 (2)	16.1 (2) (~ 20%)	16.1 (2)	-

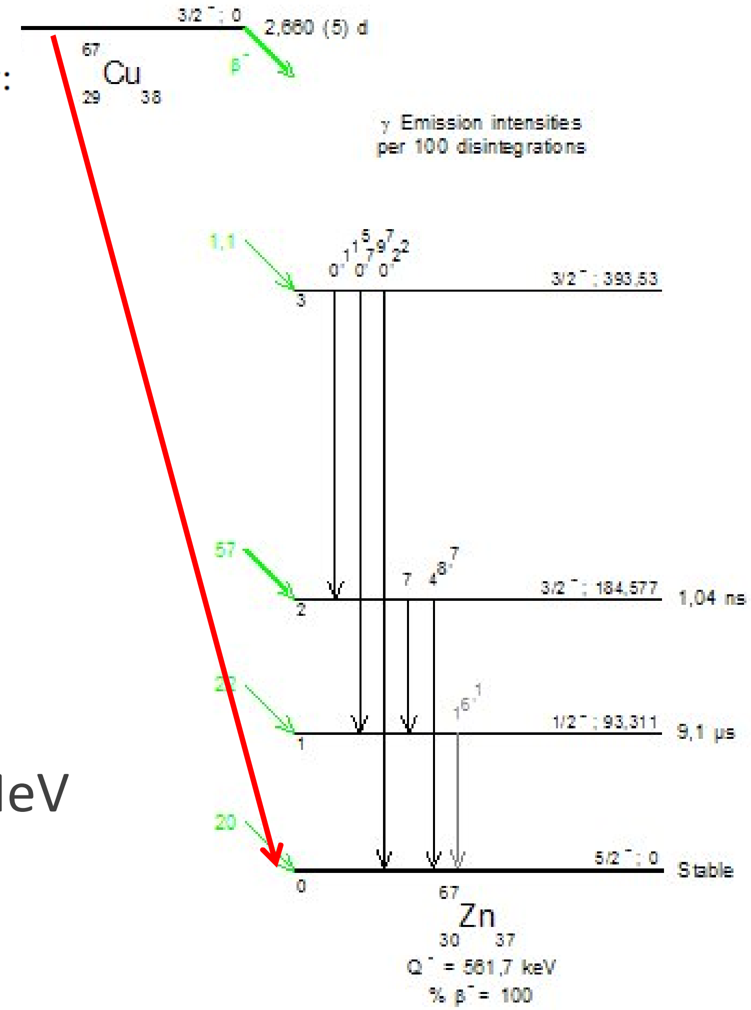
^{67}Cu – continued

Applied Radiation and Isotopes 70 (2012) 2377–2383

Review

The production, separation, and use of ^{67}Cu for radioimmunotherapy: A review

Nicholas A. Smith [a,*], Delbert L. Bowers [a], David A. Ehst [b]



Decay scheme based originally on:

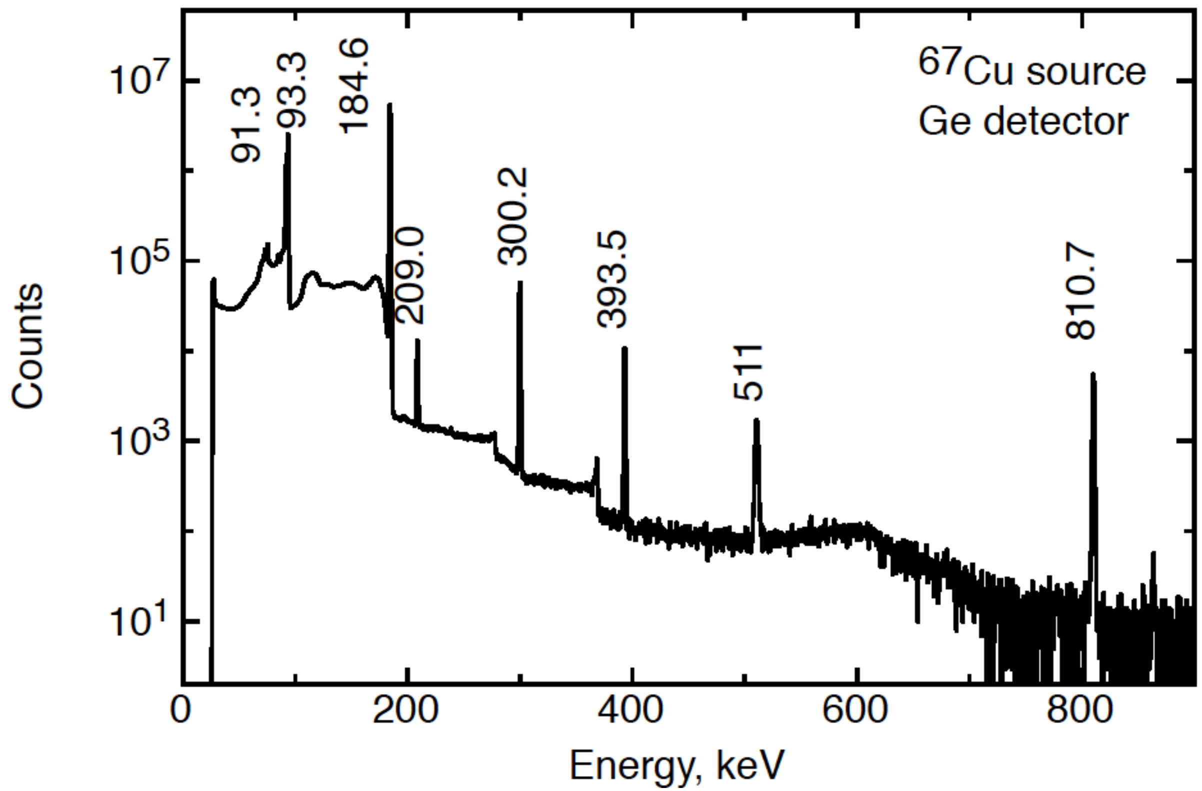
- ✓ I_β to the ground state ~ 20% (1953Ea11)
- ✓ I_γ relative values (1978Me10)

Assumption that I_β(g.s. to g.s.) is 10 (10)% translates to 11% uncertainty in production cross sections …

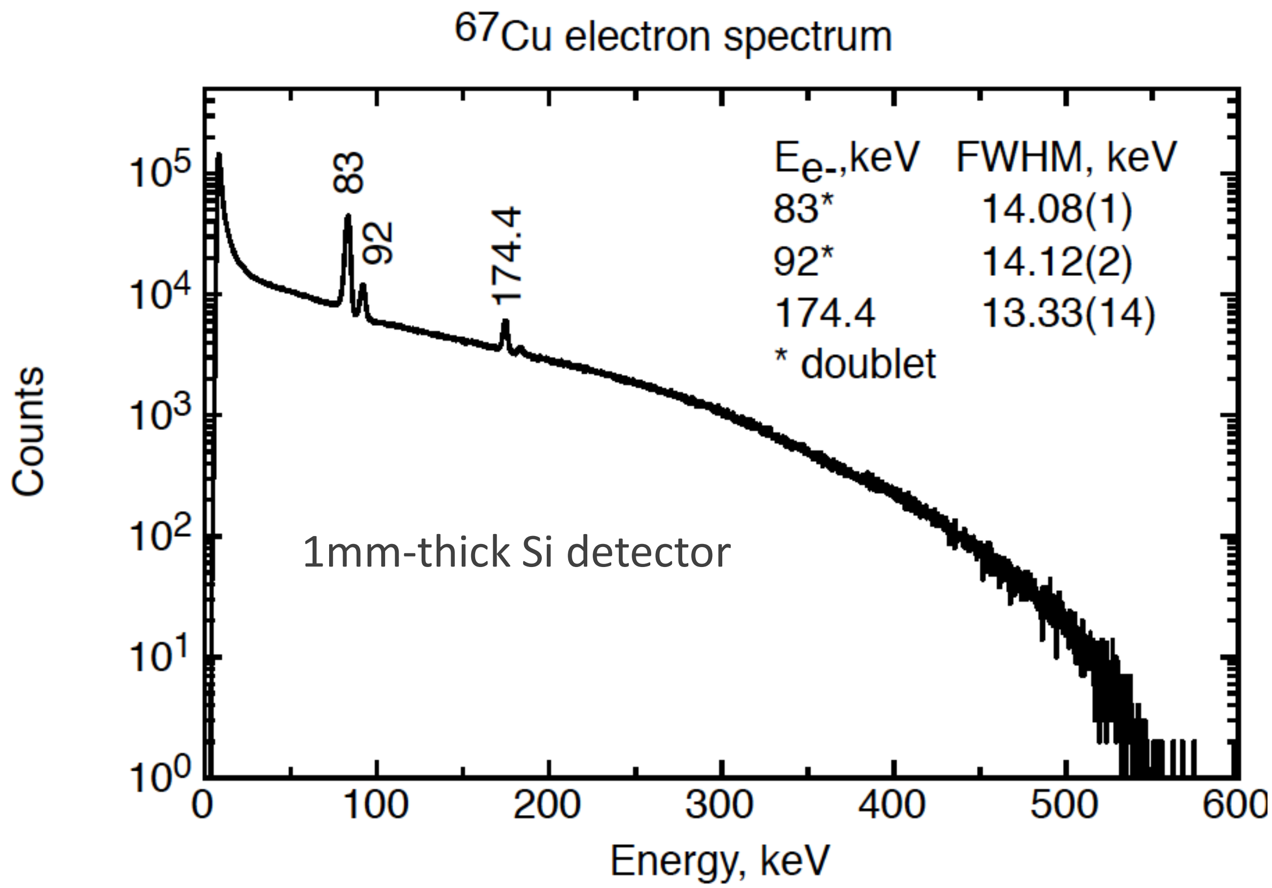
Studies at ANL:

- ✓ source produced using ^{68}Zn(γ,p)^{67}Cu, 40-MeV electron accelerator
- ✓ radiochemistry purification
- ✓ γ and β singles measurements
- ✓ βγ coincidence and correlation measurements

^{67}Cu – continued

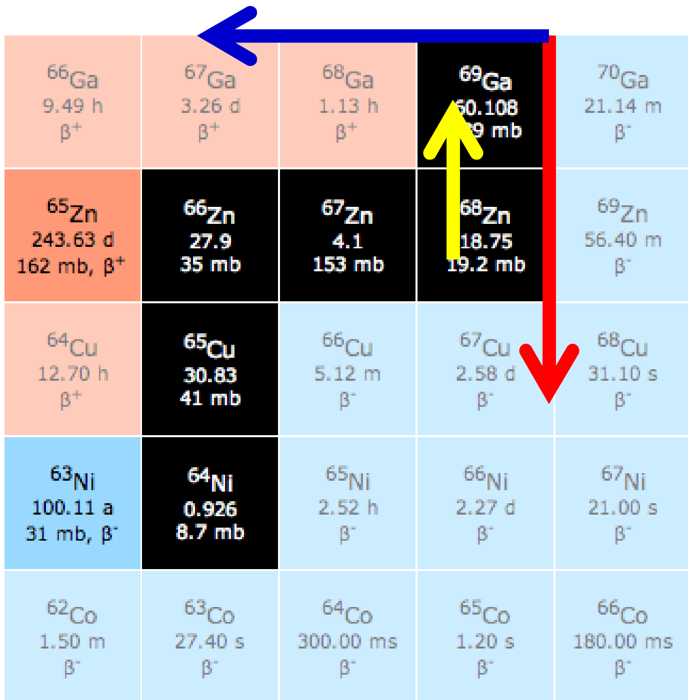


^{67}Cu – continued



Impurities/Contaminants

- ✓ depends on choice of production reaction and beam-energy regime
- ✓ depends on isotopic purity of the target



$^{68}Zn(p,2p)^{67}Cu$

$^{68}Zn(p,2n)^{67}Ga$

radiochemistry
to recycle the target

BUT …

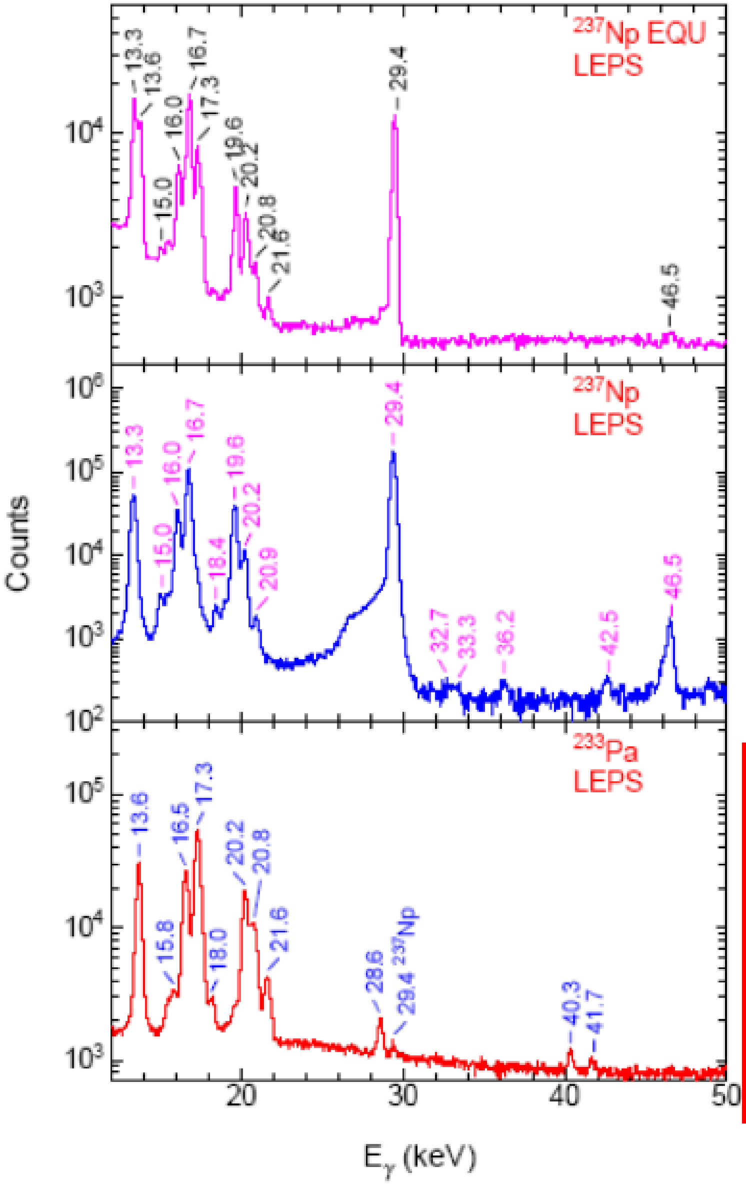
impurities are not comprehensively assessed;
require expertise in reaction paths and decay data physics

^{233}Pa γ–ray emission probabilities

purification and γ-ray measurements

- chemical separation to extract ^{233}Pa from ^{237}Np
  - dissolved ^{237}Np (in equilibrium with ^{233}Pa) in HNO_3 – solution transferred to a beaker and dried – ^{237}Np (but not ^{233}Pa) dissolved in 4 M HNO_3 – repeat procedure several times to achieve desired purity
  - several sources prepared and measured with 3-cm^3 LEPS and 25% Ge detectors – efficiency calibration determined using mixed source of ^{57,60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{113}Sn, ^{137}Cs, ^{139}Ce, ^{203}Hg and ^{241}Am, and isotopically pure ^{243}Am source – accuracy ~1% for low- and high-energy photons

^{233}Pa γ–ray emission probabilities - continued

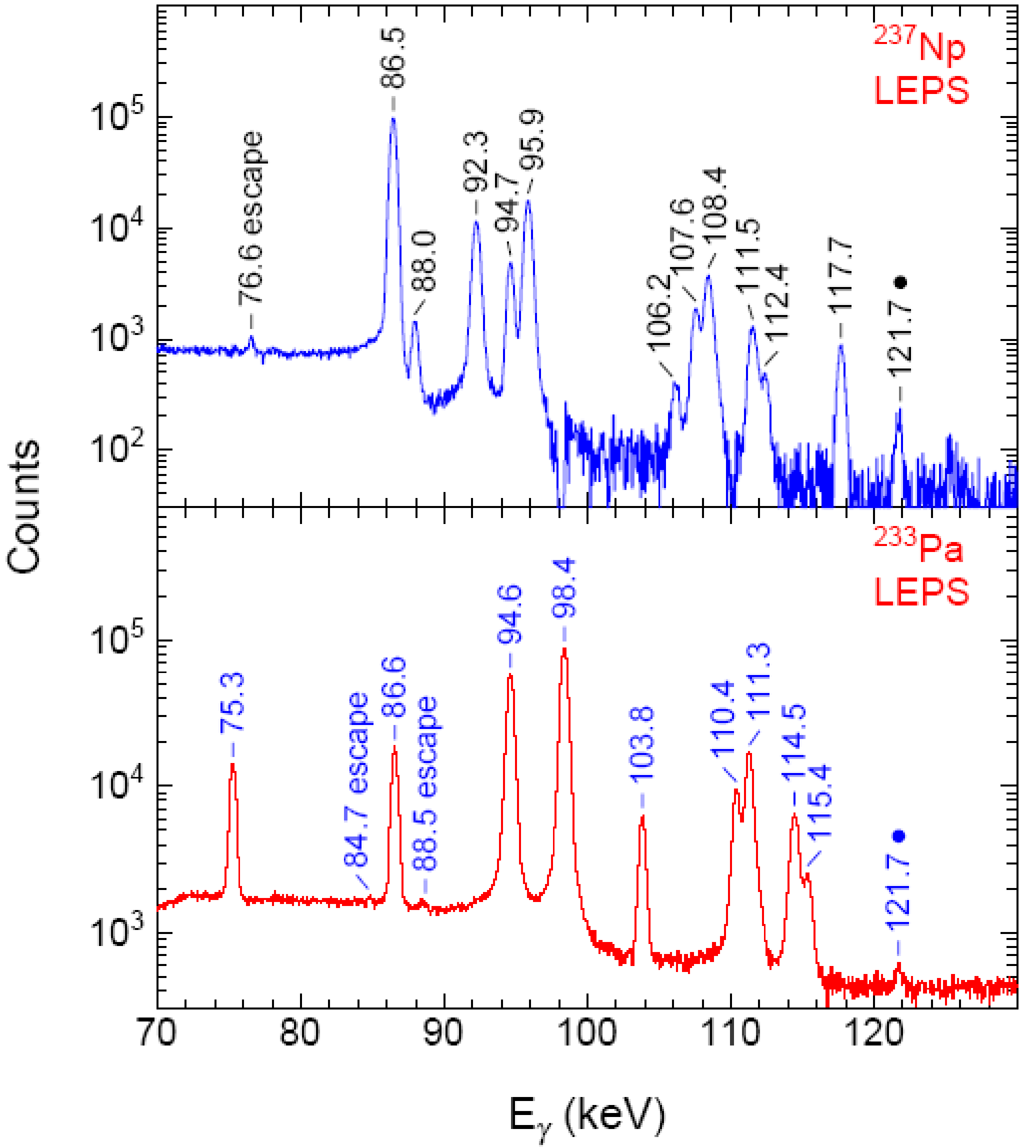


- 29.4 keV ^{237}Np line dominates – high Compton tail masks 28.6 keV (^{233}Pa)
- Compton background associated with much stronger high-energy γ rays of ^{233}Pa
- $P_\gamma(29.4/^{237}Np)/P_\gamma(75.3/^{233}Pa)$ = 10.6 (1)

- 75.3 keV line (^{233}Pa) has disappeared – pure Pa X-rays (from decay of ^{237}Np)
- Compton background associated with high-energy γ rays of ^{233}Pa is reduced

- 29.4 keV ^{237}Np line is significantly reduced, e.g. $P_\gamma(29.4/^{237}Np)/P_\gamma(75.3/^{233}Pa)$ = 0.060 (17)
- pure U X-rays
- S(28.6 keV) = 9000 counts – statistical uncertainty of about 1%

^{233}Pa γ–ray emission probabilities - continued



- resolve 86.6 keV (^{233}Pa) and 86.5 keV (^{237}Np) lines
- resolve 94.6 keV (^{233}Pa) and 94.7 keV (^{237}Np) lines
- resolve 111.3 keV (^{233}Pa) and 111.5 keV (^{237}Np) lines

ANL measurements of relevance to IAEA CRP

INDC(NDS)-0591

^{66}Ga – production via $^{66}Zn(p,n)^{66}Ga$, 12 MeV protons

^{86}Y – production via $^{86}Sr(p,n)^{86}Y$, 10-15 MeV protons

^{67}Cu

initiate a new measurement programme –
Auger emitters

Photonuclear Reactions

$^{112}Sn/^{111}In$
$^{58}Ni/^{57}Co$
$^{96}Ru/^{95}Tc^{m}$
$^{150}Nd/^{149}Pm$
$^{197}Au/^{195}Pt^{m}$
$^{70}Ge/^{68}Ge \longrightarrow {}^{68}Ga$

Production of Medical Radioisotopes with High Specific Activity in Photonuclear Reactions with γ Beams of High Intensity and Large Brilliance

D. Habs[1], and U. Köster[2]

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[2] Institut Laue Langevin, 6 rue Jules Horowitz, F-38042 Grenoble Cedex 9, France

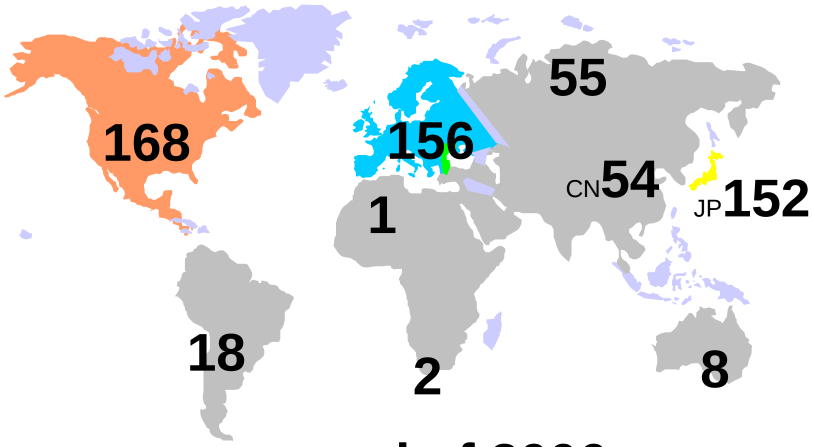
€380 M

extreme light infrastructure

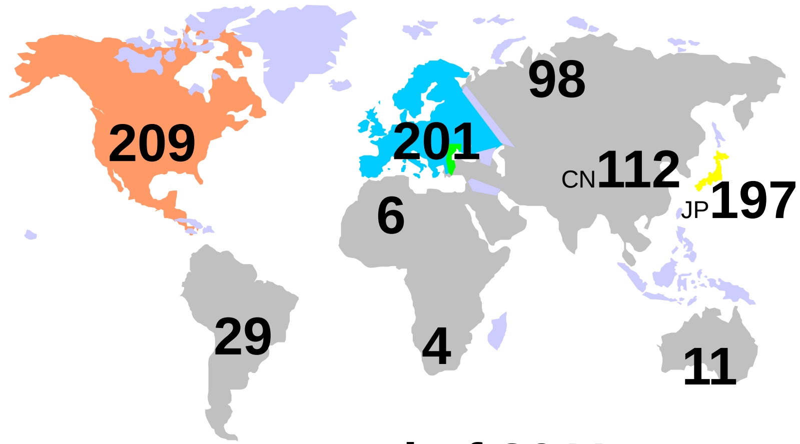
- same nuclei as with charged-particle induced reactions
- photofission $\longrightarrow {}^{99}Mo/^{99}Tc^{m}$ TRIUMF

Future opportunities

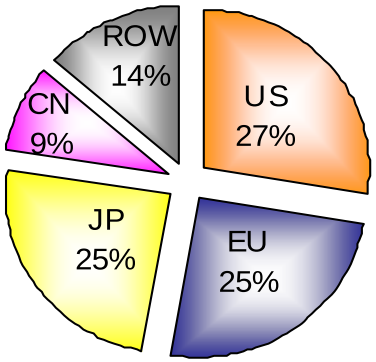
Total worldwide PET cyclotrons 2006-2011



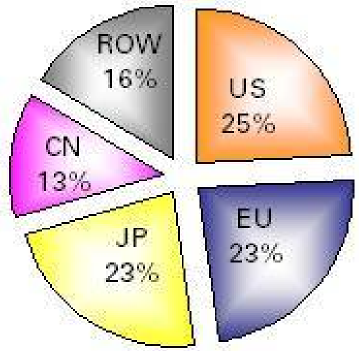
end of 2006
613 cyclotrons



end of 2011
863 cyclotrons

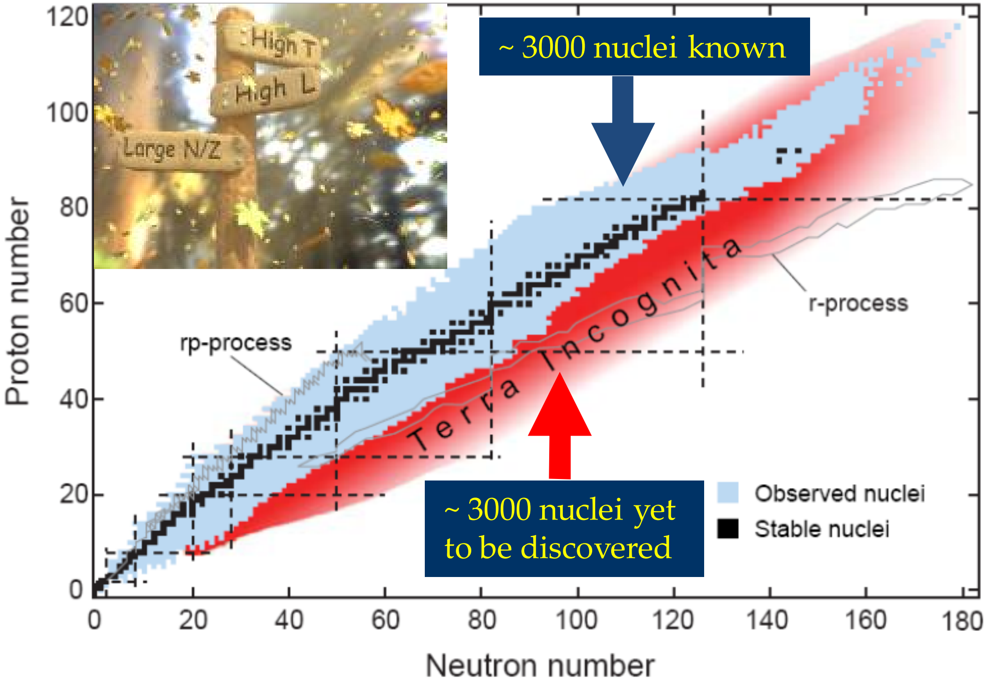


period 2007-2011
average of 50 cyclotrons
sold per year



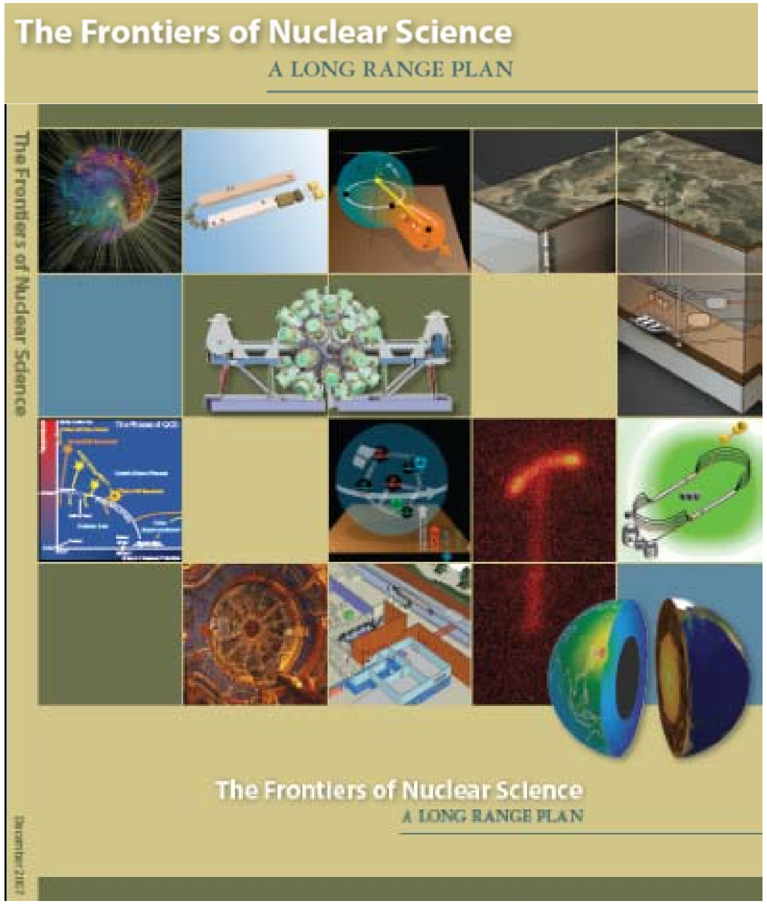
growing demands for medical isotopes

Radioactive Beam Facilities



RIKEN, TRIUMF, GANIL, CERN,
GSI (planned), RIBLL (Lanzhou), KoRIA

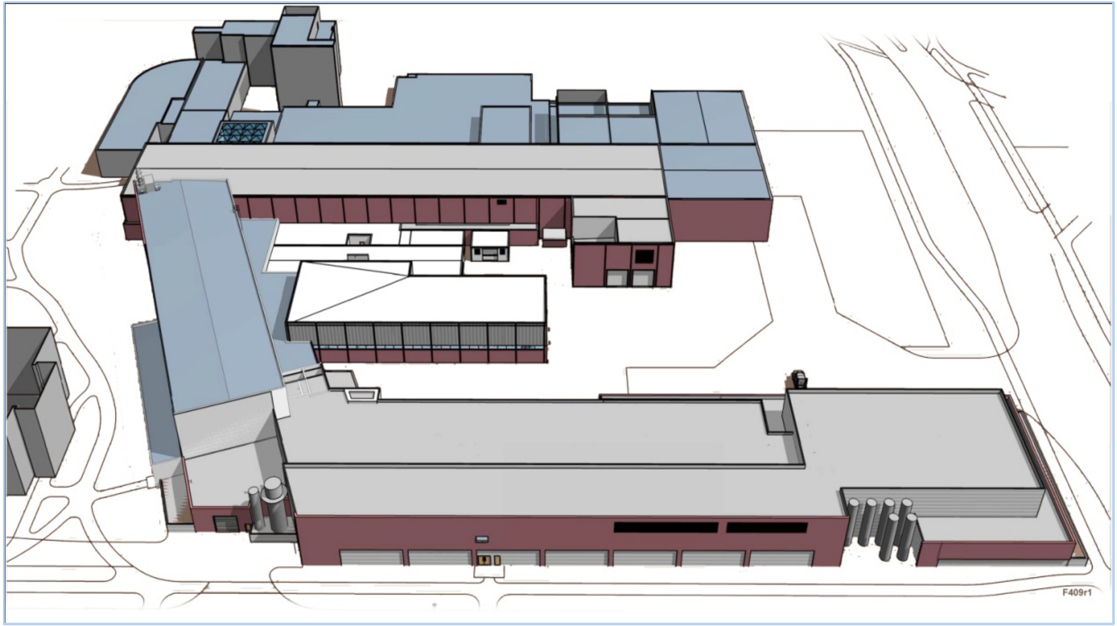
ANL, ORNL & MSU
FRIB – the future



Major New Initiative by US Community – Facility for Rare Isotope Beams (FRIB)

- Laboratory Director Konrad Gelbke; Project Director Thomas Glasmacher
- Estimated total project costs of $614.5M
- Project completion in 2020; possible early completion in 2018
- Unique key features
  - 400-kW heavy ion beams
  - efficient acceleration (multiple charge states)
  - stopped and re-accelerated separated beams
- Space for
  - re-accelerated beams, uranium up to 15 MeV/u
  - **isotope harvesting**

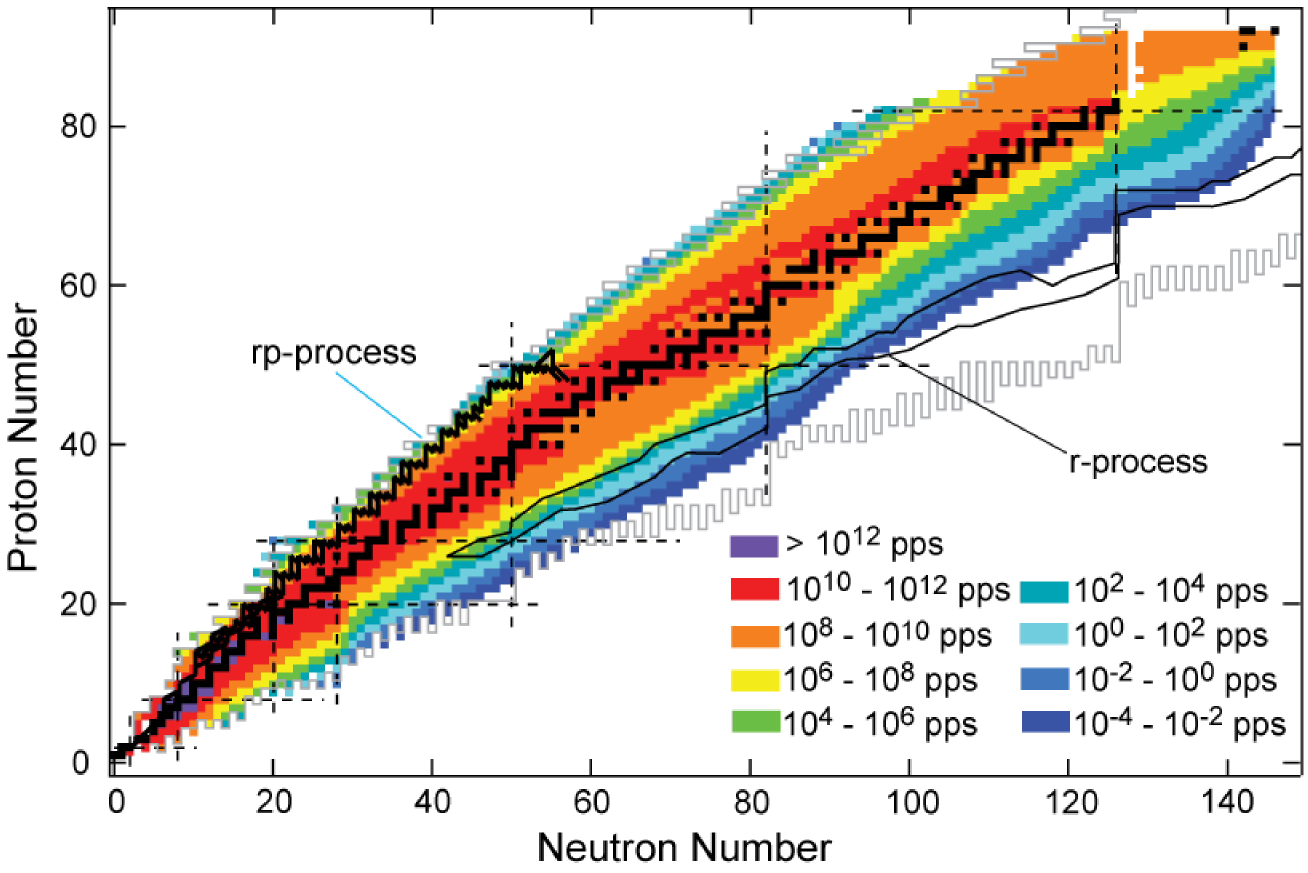
FRIB



Potential of FRIB

- FRIB will produce more than 1000 NEW isotopes at useful rates (4500 available for study, compared with 1700 now)
- Primary user at FRIB will study only one or a few isotopes – others could be harvested parasitically
- Primary beam will stop in a water beam dump; isotopes produced in the water could also be harvested.

One man's waste is another man's treasure



Rates are available at *http://groups.nscl.msu.edu/frib/rates/*

Sample Isotopes

Summary of a few selected isotopes of interest from workshop on harvesting isotopes at FRIB, Santa Fe, October 2010

Nuclide	Half-life	Use
^{32}Si	153 y	Oceanographic studies; climate change
^{221}Rn	25 m	Targeted alpha therapy
$^{225}Ra/^{229}Pa$	15 d	EDM search in atomic systems
^{48}V , ^{85}Kr	11 y	High specific activity ^{85}Kr for s-process and homeland security
^{44}Ti	60 y	Target and ion-source material
^{67}Cu	62 h	Imaging and therapy for hypoxic tumors

Selected examples

Nuclide	Primary user	Mass slits +	Near dump *	In dump #
^{28}Mg	700 mCi/day	unlikely	20 mCi/day	7 mCi/day
^{32}Si	0.063 mCi/day, 23 mCi/year	0.1 mCi/week	0.01 mCi/day	1 mCi/year
^{44}Ti	10 mCi/year	0.1 mCi/week	0.1 mCi/year	0.1 mCi/year
^{67}Cu	2000 mCi/day	100 mCi/day	100 mCi/day	100 mCi/day
^{99}Mo				1500 mCi/day

+ Collection at slits might be available one week per year

* Near dump rates are an approximate average of production from favorable beams on the beam list, available around 40 days per year

Assumes ^{238}U primary beam available around 150 days per year; higher yields possible from other primary beams

IAEA-NDS Medical Portal

Repository for and dissemination of nuclear data for medical applications:

IAEA-NDS Medical Portal must be the focal point

http://www-nds.iaea.org/medportal/

α, β^+, γ and Auger-electron Decay Data in Nuclear Medicine − Experimental Determination, Status and Deficiencies

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Workshop on Nuclear Data for Science and Technology: Medical Applications
ICTP, Trieste, Italy, 30 September – 4 October 2013