

70 YEARS OF CREATING TOMORROW



**Los Alamos**  
NATIONAL LABORATORY

# Parasitic Isotope Production with Cyclotron Beam Generated Neutrons

*J.W. Engle, E.R Birnbaum, M.E. Fassbender,  
K.D. John and F.M. Nortier*

LANL Isotope Program  
Chemistry Division

UNCLASSIFIED



# Accelerator production of isotopes

TABLE 3.2. POPULAR RADIONUCLIDES VERSUS THE PROTON ENERGIES REQUIRED FOR THEIR PRODUCTION

Proton energy (MeV)	Radionuclides easily produced
0–10	F-18, O-15
11–16	C-11, F-18, N-13, O-15, Na-22, V-48
17–30	I-124, I-123, Ga-67, In-111, C-11, F-18, N-13, O-15, Na-22, V-48, Tl-201
30+	I-124, I-123, Ga-67, In-111, C-11, F-18, N-13, O-15, Sr-82, Ge-68, Na-22, V-48

Intermediate energies  
~60 MeV and higher

IAEA Technical Report Series No. 465, 2008



# Operating Intermediate Energy Facilities Worldwide

- LANL, USA – 100 MeV, 250 µA
- BNL, USA – 200 MeV, 100 µA
- INR, Russia – 160 MeV, 120 µA

{



- iThemba, South Africa – 66 MeV, 250 µA
- PSI, Switzerland – 72 MeV, 100 µA
- TRIUMF, Canada – 500, 70 MeV, 100 µA
- ARRONAX, France – 70 MeV, 2X 375 µA

{



From a production volume perspective  
the IPF at LANL is presently the leading  
high power facility



# Operating Intermediate Energy Facilities Worldwide

Facility	Beam Energy (MeV)	Operating Beam Current ( $\mu$ A)
ARRONAX, France	70	2X 375
LANL, USA	100	250
iThemba, South Africa	66	250
INR, Russia	160	120
BNL, USA	200	100
TRIUMF, Canada	500, 70	100
PSI, Switzerland	72	100

New Facility	Beam Energy (MeV)	Beam Current ( $\mu$ A)
Deajeon, Korea	100	600



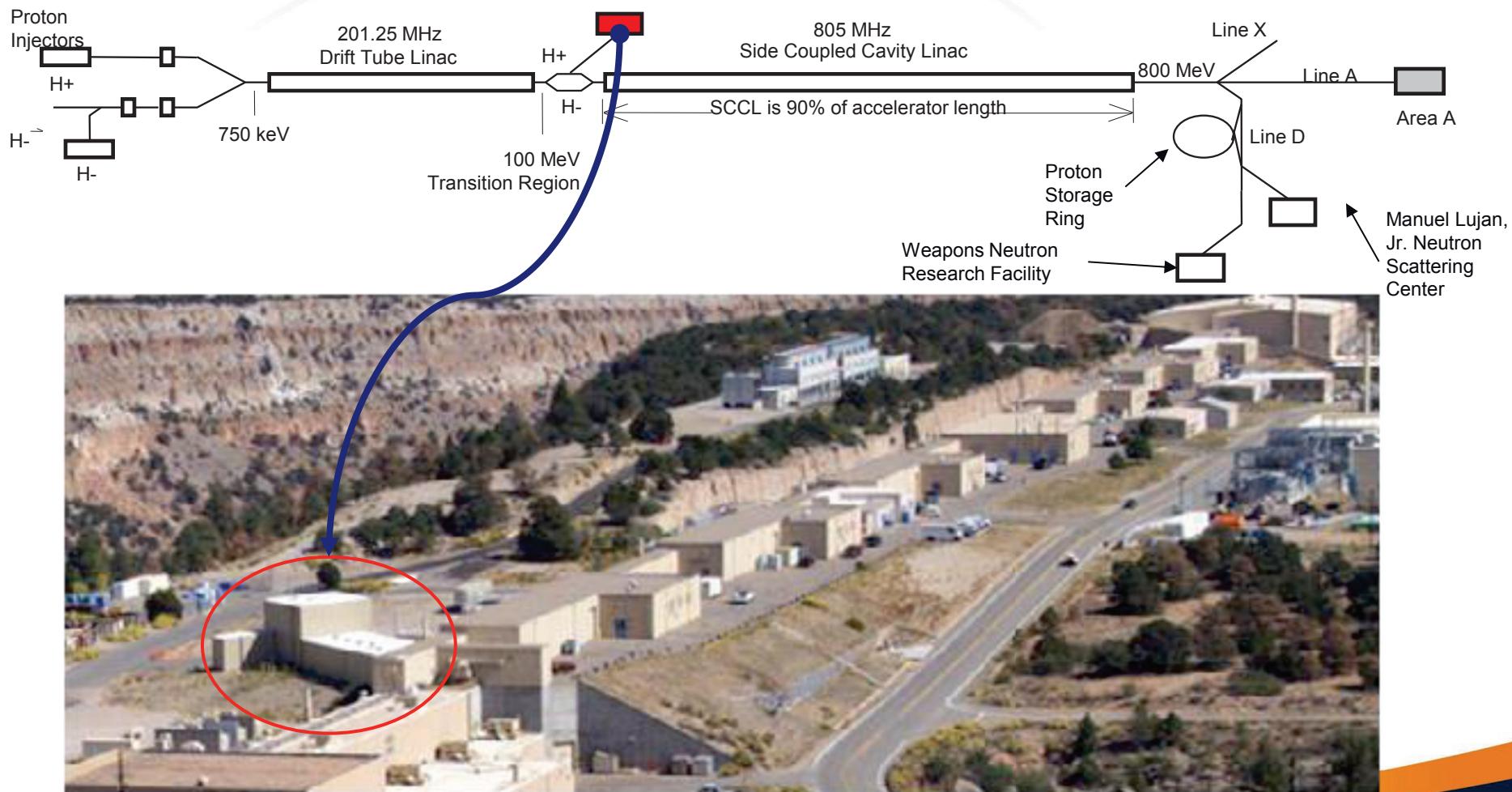
# The LANL experience – commercial isotopes $^{82}\text{Sr}$ and $^{68}\text{Ge}$ produced on a large scale

Isotope	Half-life	Main Use
$^{82}\text{Sr}$	25.5 d	Parent of $^{82}\text{Rb}$ used in cardiac perfusion studies with PET
$^{68}\text{Ge}$	270 d	Positron emitter used in calibration sources for every PET scanner in clinical use
$^{88}\text{Zr}$	83.4 d	Parent of $^{88}\text{Y}$ used as a tracer surrogate for $^{90}\text{Y}$ in oncological bio-distribution studies
$^{22}\text{Na}$	2.6 a	Test objects for PET studies, tracer for natural Na
$^{32}\text{Si}$	153 a	Tracer for environmental transport studies
$^{73}\text{As}$	80.3 d	Tracer for toxicology studies
$^{109}\text{Cd}$	462.6 d	Source for X-ray fluorescence
$^{67}\text{Cu}$	2.6 d	Treatment of non-Hodgkin's Lymphoma
$^{225}\text{Ac}$	10 d	Alpha emitter used in cancer therapy clinical trials
$^{186\text{g}}\text{Re}$	90.6 h	Bone pain palliation, cancer therapy

Other facilities has similar experience



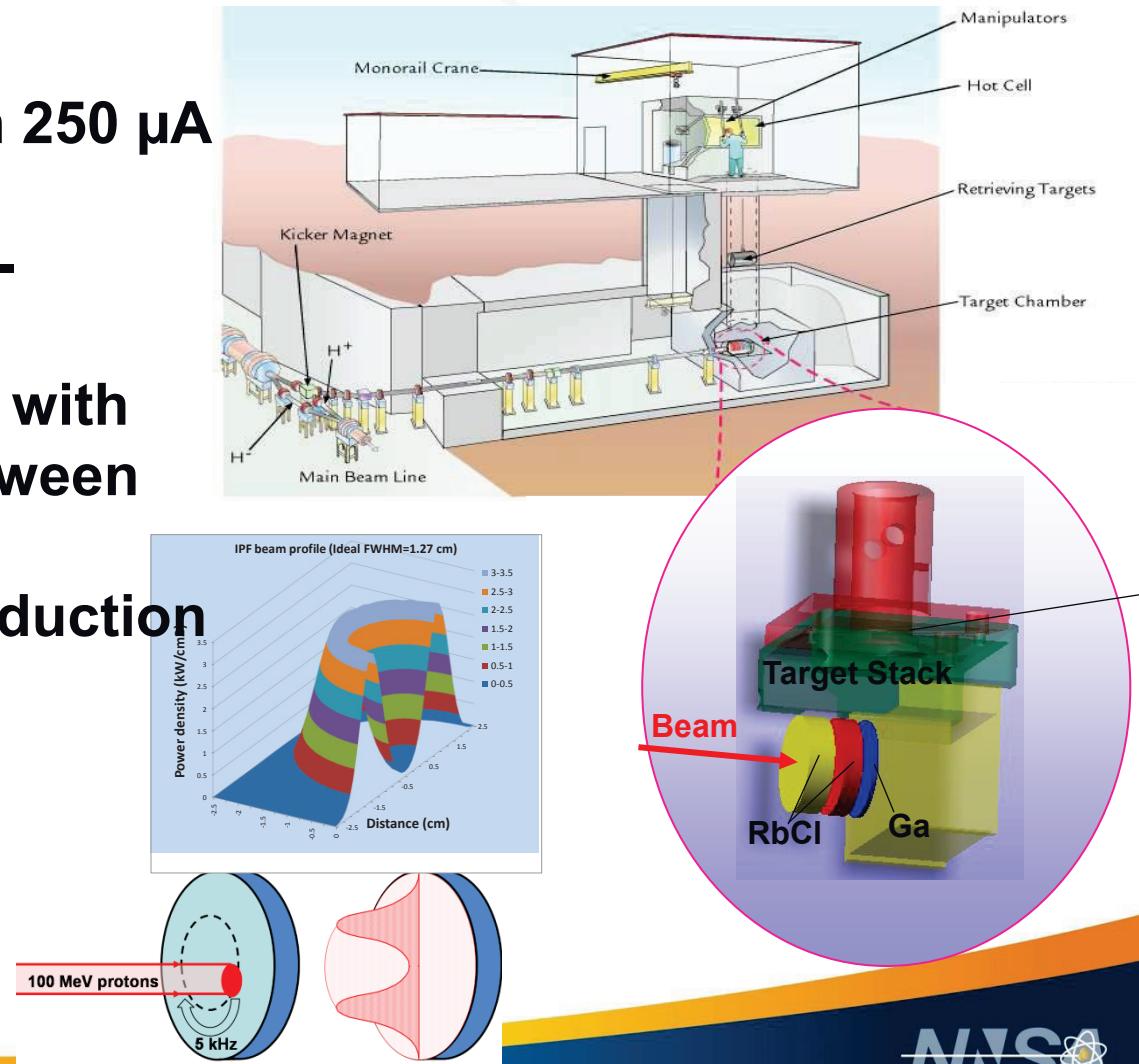
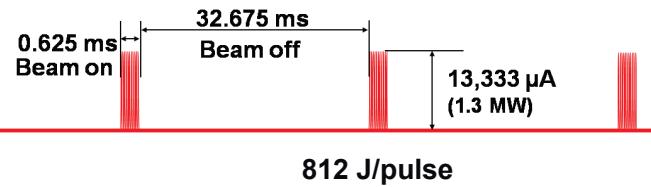
# Overview – Isotope Production Facility (IPF) at LANSCE





# Overview - Large Scale Production

- Production targets are routinely irradiated with 250  $\mu\text{A}$  of 100 MeV protons
- Pulsed beam has a ring-shaped profile
- Three targets in a stack with cooling channels in between
- Production occurs simultaneously in 3 production energy windows



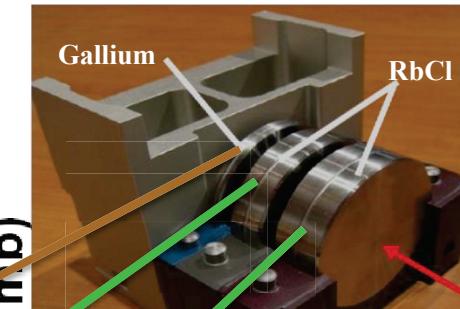


# One Target Stack Tuned for $^{82}\text{Sr}$ and $^{68}\text{Ge}$

(mb)

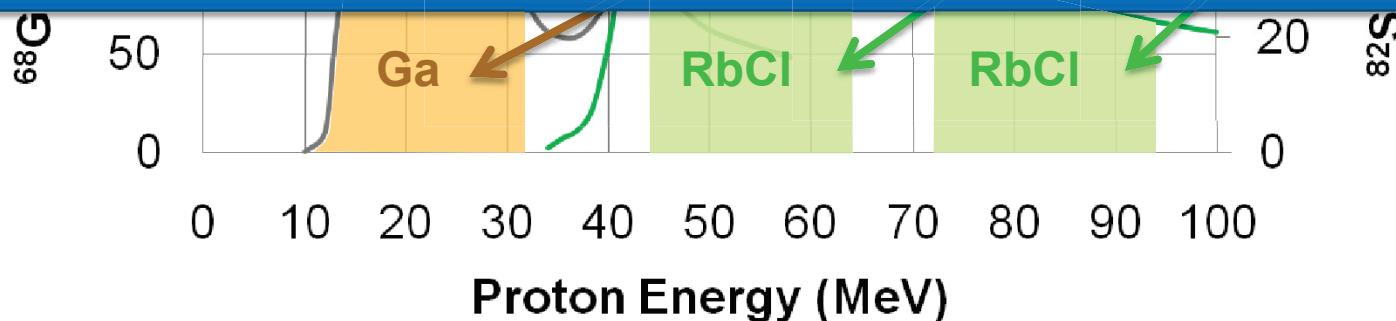


(mb)



Represents majority target configuration  
for IPF

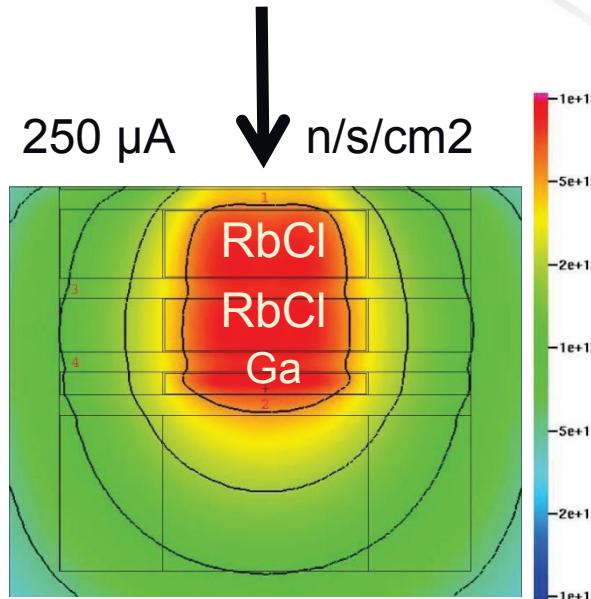
Challenge to allocate beam time to research and new  
isotopes





# Lots of Secondary Neutrons

MCNPX prediction



“nuisance” neutrons cause  
unwanted activation  
unwanted production of  
impurities

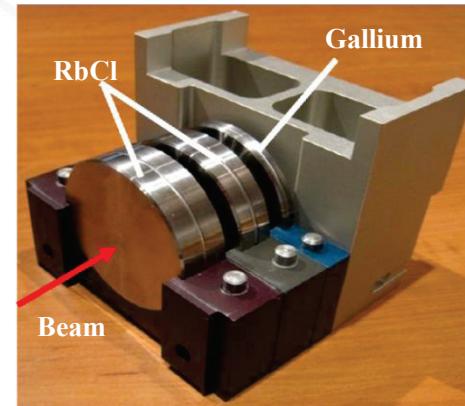
- For the  $^{82}\text{Sr}/^{68}\text{Ge}$  production target stack
- Model calculations predict a secondary neutron flux of  $\sim 10^{12} \text{ n/s/cm}^2$  around the targets

Comparable with that of a medium-flux  
research reactor



# Parasitic Production of NCA Isotopes Utilizing Secondary Neutrons

- Presents potential for useful production of No-Carrier-Added isotopes via (n,p), (n,2n) and (n, $\alpha$ ) threshold reactions
- Candidate isotopes include  $^{36}\text{Cl}$ ,  $^{63}\text{Ni}$ ,  $^{64}\text{Cu}$ ,  $^{67}\text{Cu}$ ,  $^{85}\text{Kr}$ ,  $^{89}\text{Zr}$ ,  $^{212}\text{Pb}$ ,  $^{225}\text{Ac}$ ,  $^{229}\text{Th}$ ,  $^{231}\text{Pa}$ ,  $^{237}\text{Np}$
- Inspection of reaction thresholds on target nuclei over a wide mass range shows that thresholds generally vary in the range 0-10 MeV

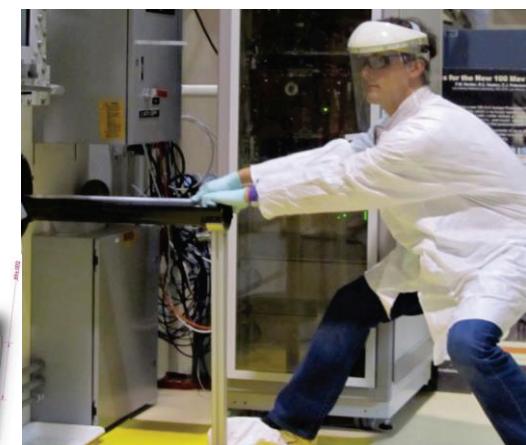
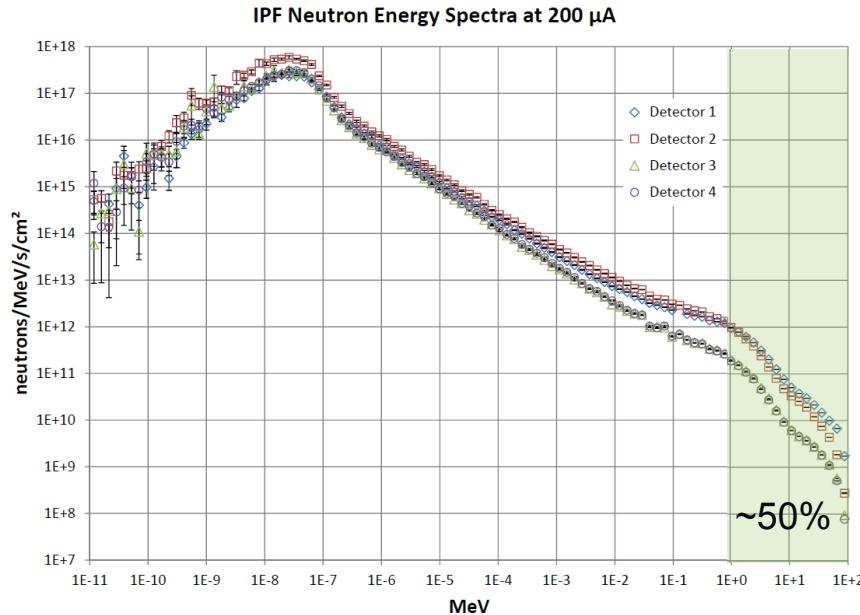


Target Nucleus	(n,p)	(n,2n)	(n, $\alpha$ )
$^{16}\text{O}$	10.2	16.6	2.3
$^{36}\text{Cl}$	0	8.8	0
$^{70}\text{Zn}$	5.8	9.3	0.2
$^{148}\text{Nd}$	4.1	7.4	0
$^{226}\text{Ra}$	2.9	6.4	0



# Parasitic Production of NCA Isotopes Utilizing Secondary Neutrons

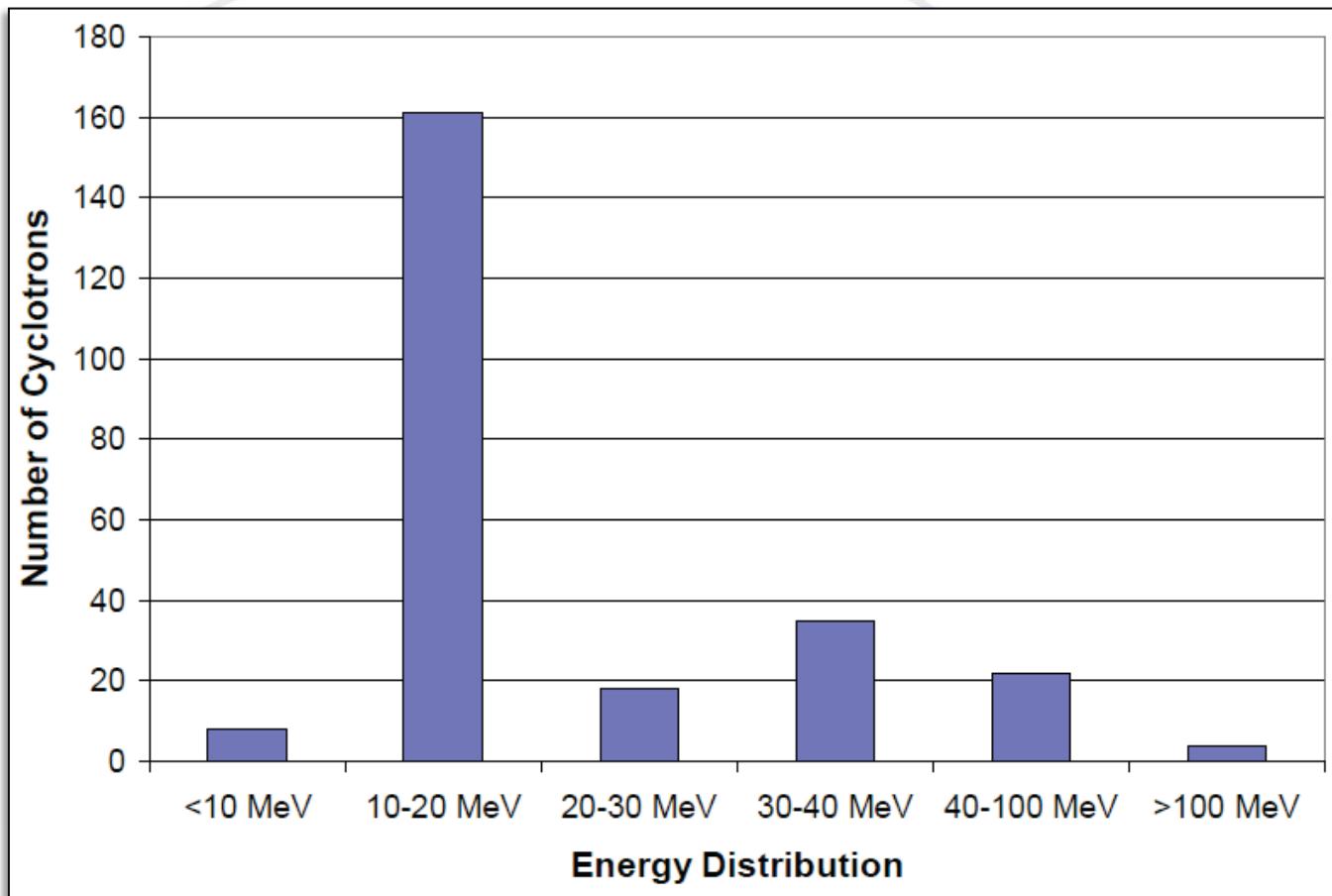
- Approximately 50% of the neutrons have energies  $> 1 \text{ MeV}$  (25%  $> 5 \text{ MeV}$ , 15%  $> 10 \text{ MeV}$ )
- This high energy component is very suitable for inducing  $(n,p)$ ,  $(n,2n)$  and  $(n,\alpha)$  type reactions
- MCNPX model calculations and experimental verification measurements suggests small scale production is feasible
- Work is championed by Post Doctoral Fellow, Jonathan Engle



J.W. Engle et al., AIP Conf. Proc., 1509 (2012)



# 2006 - Cyclotrons energy distribution



IAEA - DCRP, 2006

Intermediate energy cyclotrons can take advantage



# Commercial 70 MeV cyclotrons for large-scale production

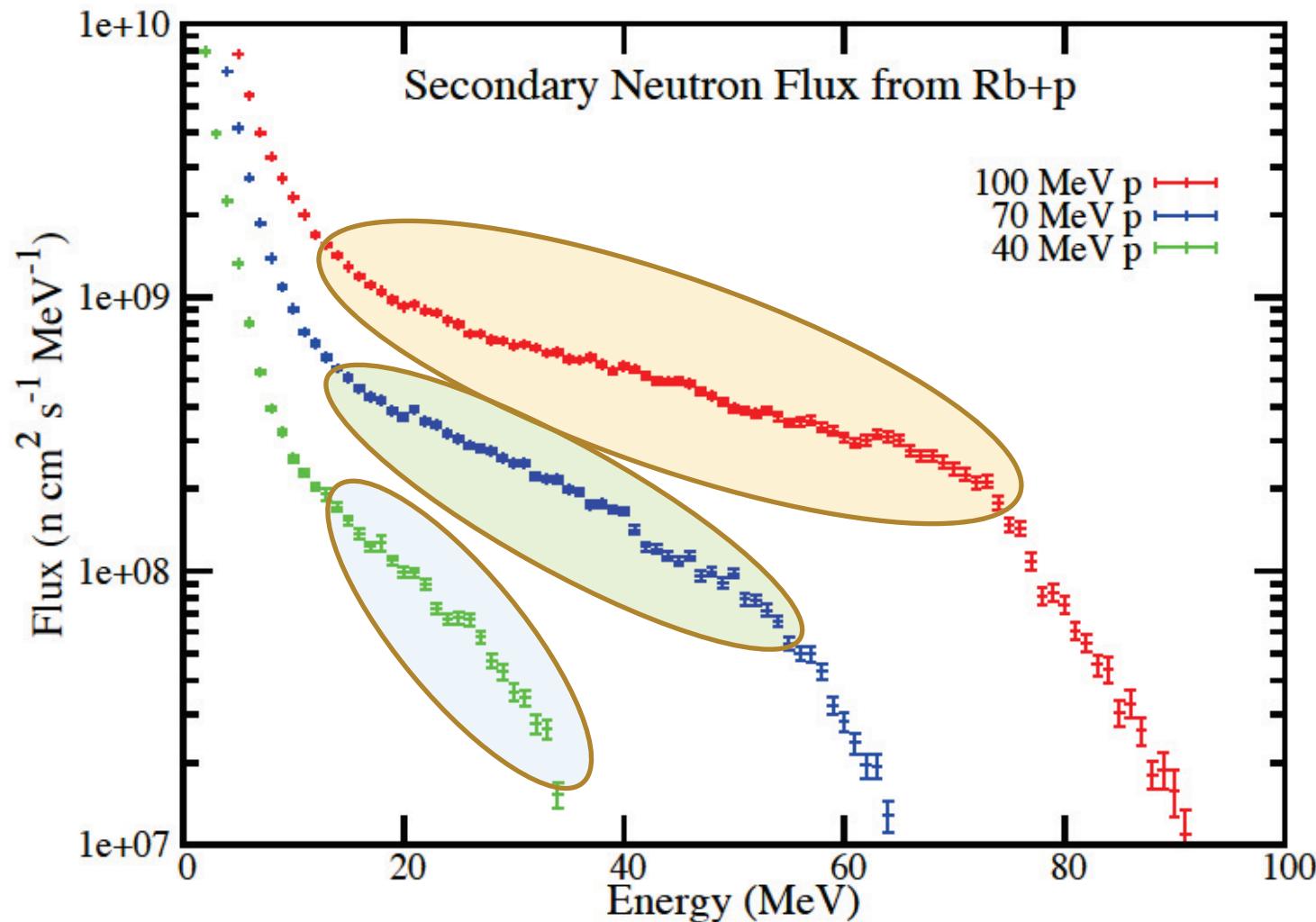
ARRONAX machine, France



Beam	Particle	Energy (MeV)	Current ( $\mu\text{Ae}$ )	Dual beam
Proton	H-	30-70	<375	Yes
	HH+	17.5	<50	No
Deuteron	D-	15-35	<50	Yes
Alpha	He++	68	<70	No

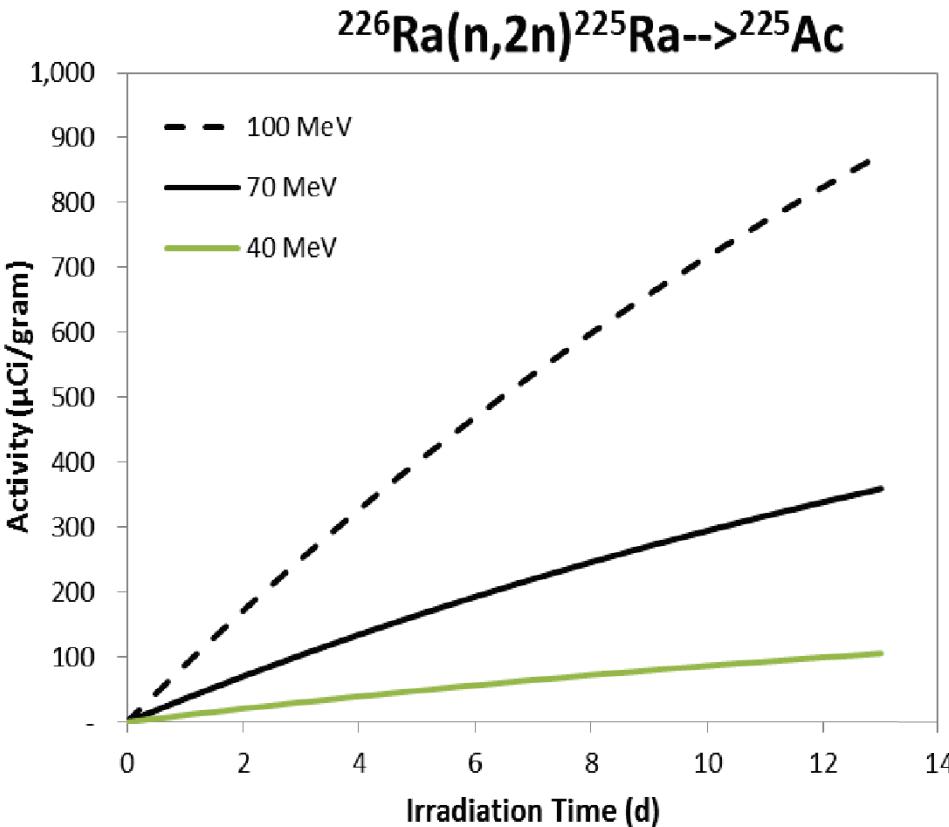


# Secondary neutron flux comparison





# Example of small-scale production of NCA $^{225}\text{Ac}$



- Production approaching mCi scale is possible at 100 MeV, 250  $\mu\text{A}$
- Sufficient to support pre-clinical research efforts
- 70 MeV and 40 MeV potential is lower but
- High current of emerging commercial machines can still take advantage



# Advantages are compelling

- Production is parasitic with no impact on proton beam schedule
- No proton beam heating of targets
  - Allows a wide range of target material
  - Containment of target material is greatly simplified
- Recycling of expensive highly enriched target material is much simpler
  - Target mass can range from *mg* to *g* scale and beyond
  - No sophisticated hot cell based target fabrication capability required



# Summary

- Several isotope production facilities are operating at intermediate energies, including high power cyclotrons
- Often primary beam time is consumed for production of one or two isotopes (Sr-82, Ge-68 at LANL)
- Challenge to allocate beam time for small quantities of research isotopes
- At LANL parasitic utilization of secondary neutrons shows promise for research scale production via threshold reactions such as (n,p), (n,2n) and (n,α)
- This approach can benefit other facilities, especially high power cyclotron facilities such as iThemba, ARRONAX and TRIUMF



# Acknowledgements

- Modeling and experimental work performed by Jon Engle (*Distinguished Reines Fellow*)
- Colleagues in the Isotope Program (co-authors *J.W. Engle, E.R Birnbaum, M.E. Fassbender and K.D. John*)
- Funding support
  - LANL (3 year Reines Fellowship Grant)
  - DOE Office of Science, Office of Nuclear Physics

