Development of Radioisotopes with ADS Systems

Photons, Neutrons and Photonuclear Physics in isotope production

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Questions to be addressed:

- Why ADS for isotope production? Gammas, Fast Neutrons, Thermal Neutrons
- What are the “low-hanging fruit” of ADS Isotope Production? Is it Photonuclear Production?
- What role do or should high power electron linacs play?
- Are there “unique” or particularly advantageous opportunities in electron-linac ADS isotope production?

ADS, cast in its most general sense, is an accelerator driven system to produce photons, neutrons, fissions (nuclear reactions) and energy.

The (non-traditional) concept of ADS considered here is:

Zeroth Order	=	Accelerator-driven, high-brightness photon and fast neutron source (no fuel) for isotope production, burn up of nuclear waste, materials irradiations studies, with or without neutron moderation.
First Order	=	Accelerator-driven single-stage subcritical system with neutron multiplication for; isotope production, materials studies.
2nd Order	=	Accelerator-driven coupled-stages subcritical system with neutron multiplication for; power i.e. process heat etc.

Primary Applications for each "Order" of ADS with **Electron Linacs**:

Zeroth Order	=	Isotope Production (photo-nuclear +/or n-capture), Radiation Damage (fission-spectrum fast neutrons) Waste Transmutation (^{99}Tc, ^{129}I)
First Order	=	Isotope Production, (esp. via n-capture and n-fission) "Breed" ^{233}U, ^{3}H, etc. Waste Transmutation
2nd Order	=	Energy/Power (direct heat or electrical) Isotope Production "Breed" ^{233}U, ^{3}H, etc. Waste Transmutation

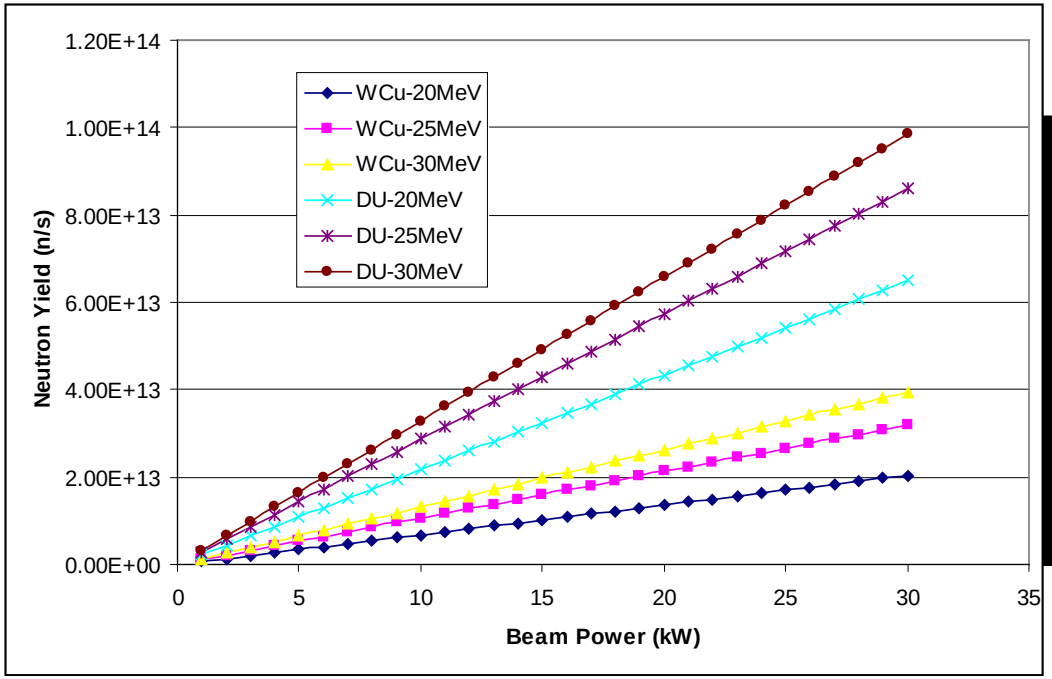
Zeroth order ADS:
a bright neutron source for materials irradiation:

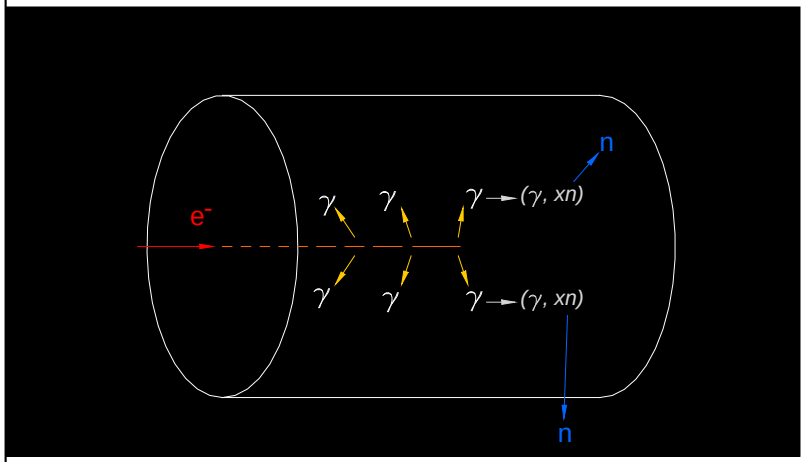
How intense and how reliable can such an electron linac-driven source be?

Because medical isotope production REQUIRES reliability.

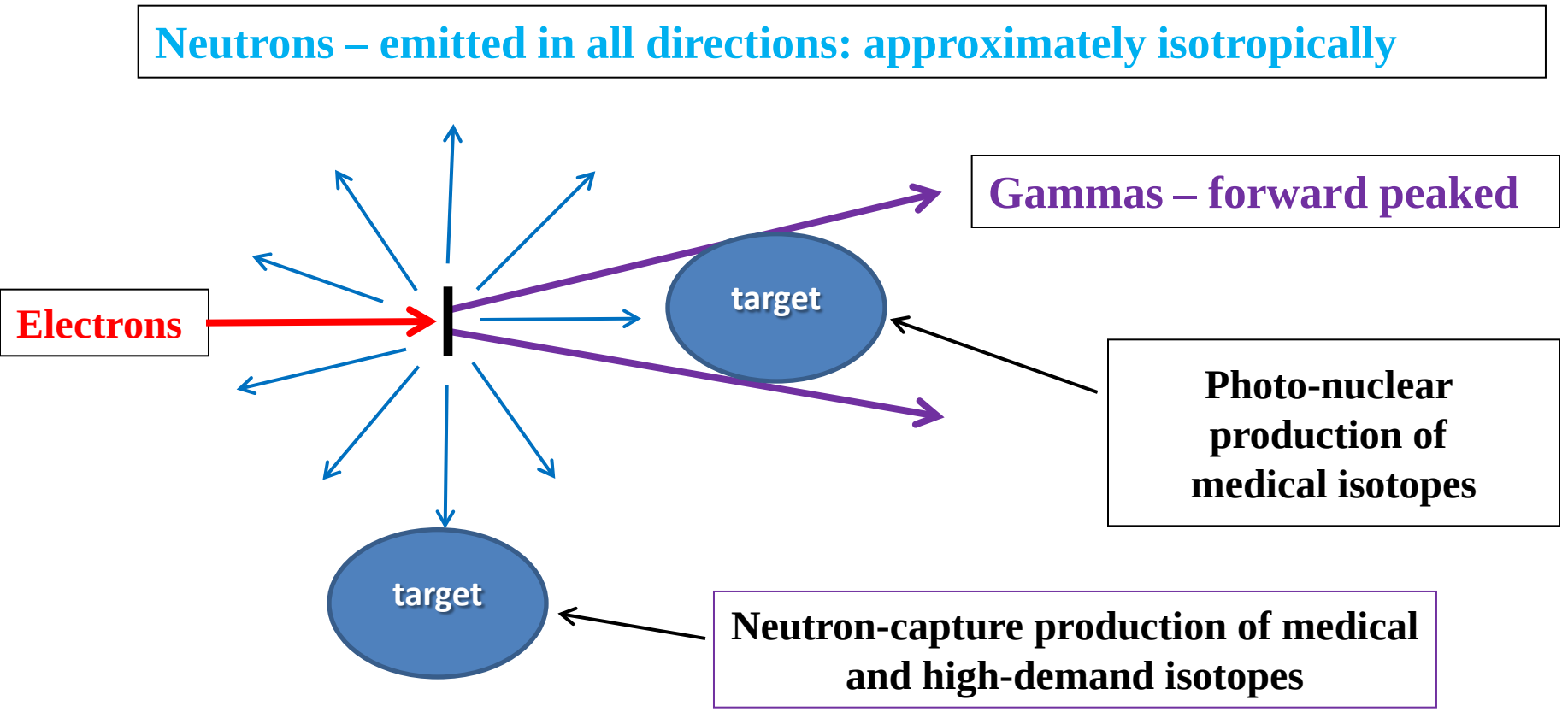
Why Zeroth order, electron-linac ADS? Neutron Source for Materials Irradiation, Waste Burnup and **Isotope Production**

A relatively low-cost fast fission-spectrum neutron source for long term irradiation of small samples is possible. A 250 kW, 100 MeV electron source could provide an inexpensive domestic fast-neutron irradiation capability for small samples (≈ few cm3) with a flux of up to ≈10^{15} n/sec (≈10^{14} n/cm^2-sec), to meet needs of fuel-cycle materials researchers



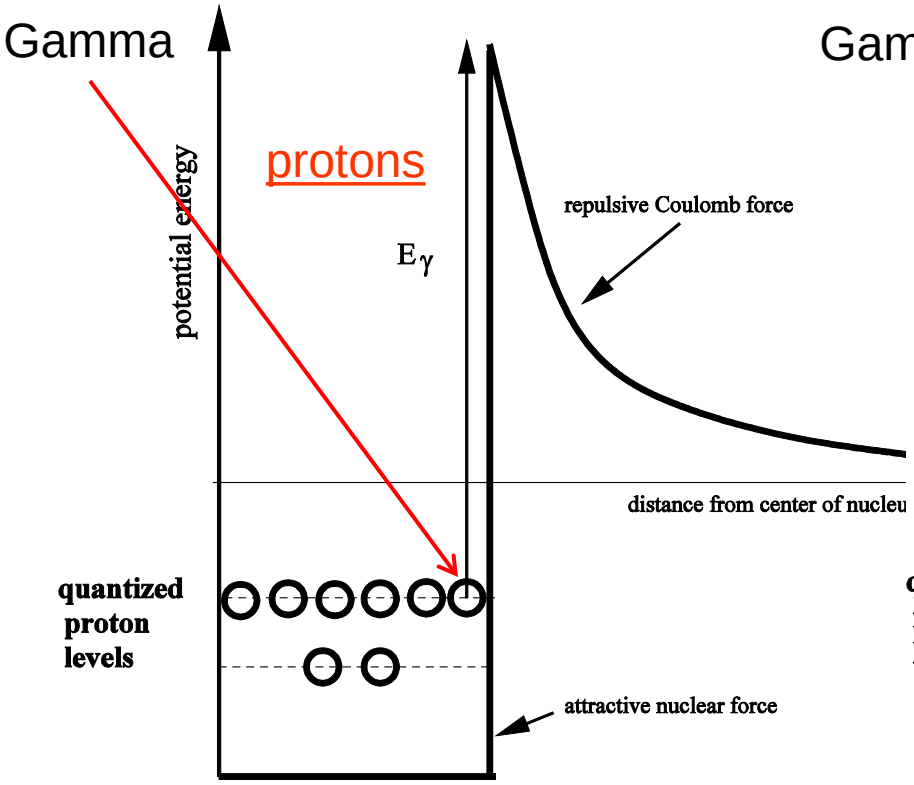


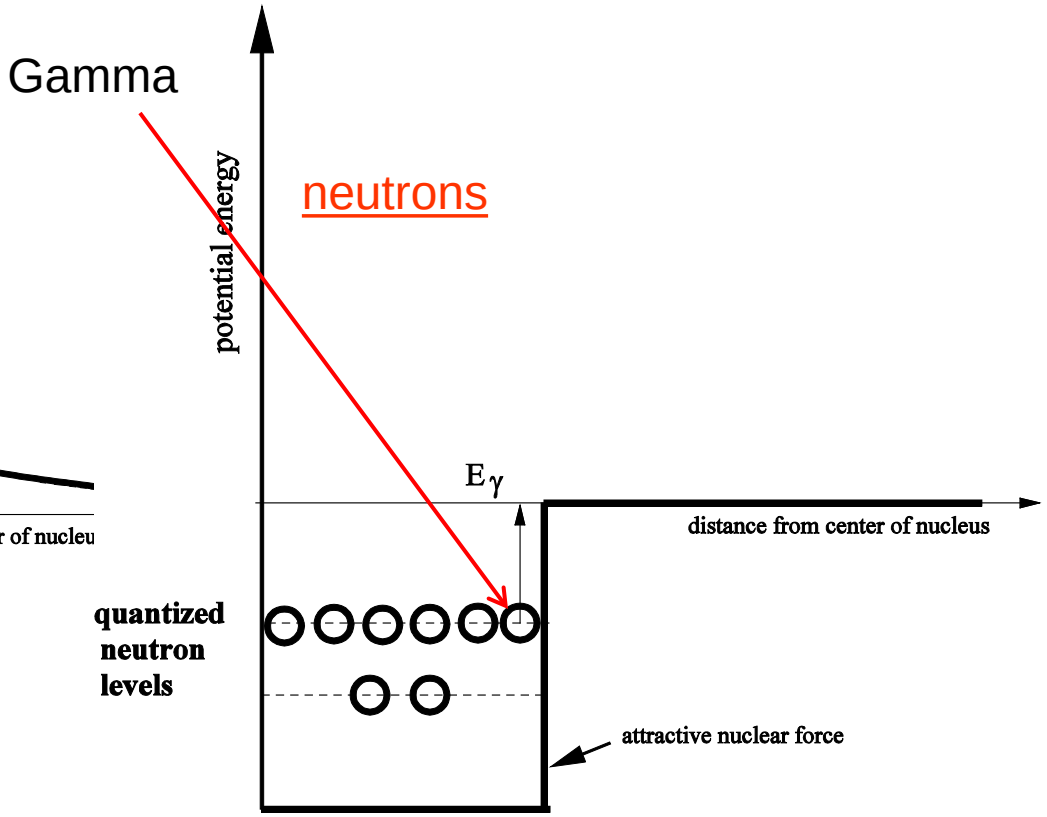
Zeroth order ADS:
a bright neutron/gamma source for isotope production



A 100 kW, 100 MeV electron linac is capable of producing 100% or the U.S. demand for many high-priority research isotopes for medical and other kinds of research. Such a device could also produce nearly 10% of the entire U.S. demand for ^{99}Mo.

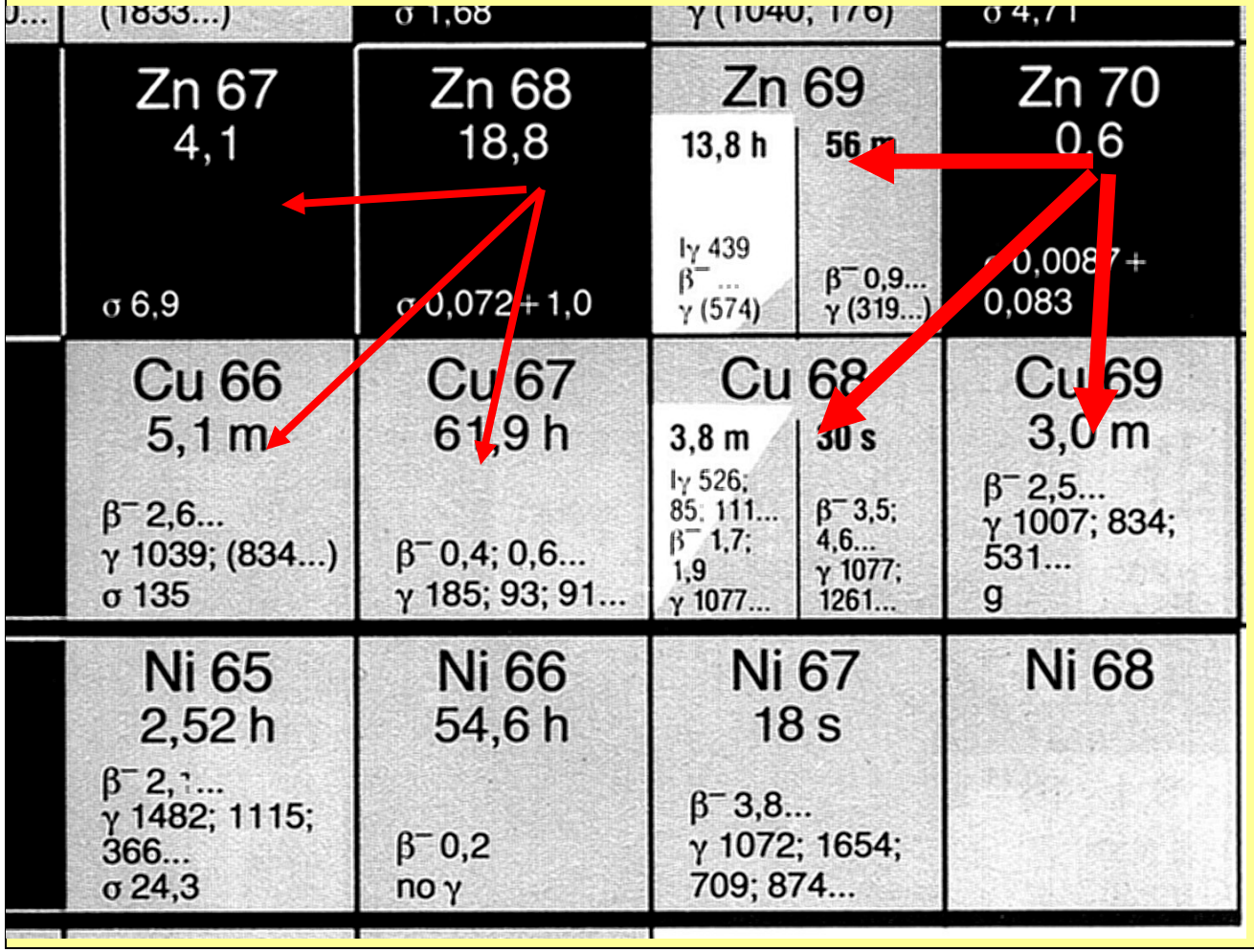
Why Photonuclear? Photons (gammas) are absorbed by the nucleus which, in turn, typically "equilibrates" (multi-nucleon excitation) before emitting particle(s) to de-excite: favors neutron emission for metals (medium to high atomic number)





Coulomb energy barrier for protons … **but not for neutrons.**

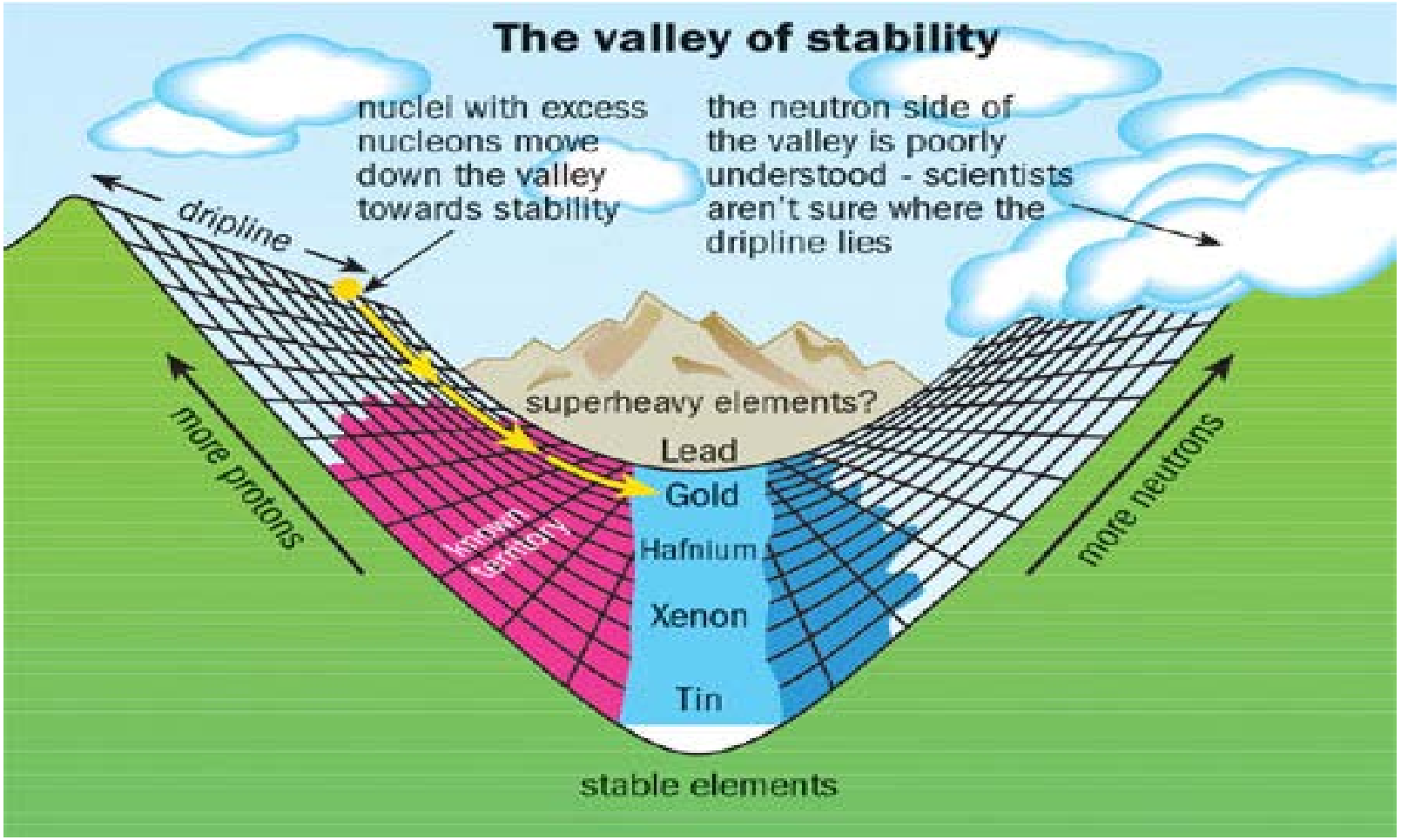
(gamma, n) and (gamma, p) reactions both play important roles, depending on the target, as do smaller cross-section reactions:
Example - Zinc Analysis:



Reaction types:

- γ, γ´
- γ, xn
- γ, p
- γ, np
- γ, α
- γ, αn
- γ, f
- γ, ...

Photo-nuclear reactions enable fuller exploitation of the moderately Proton-Rich side of the Valley of Stability …

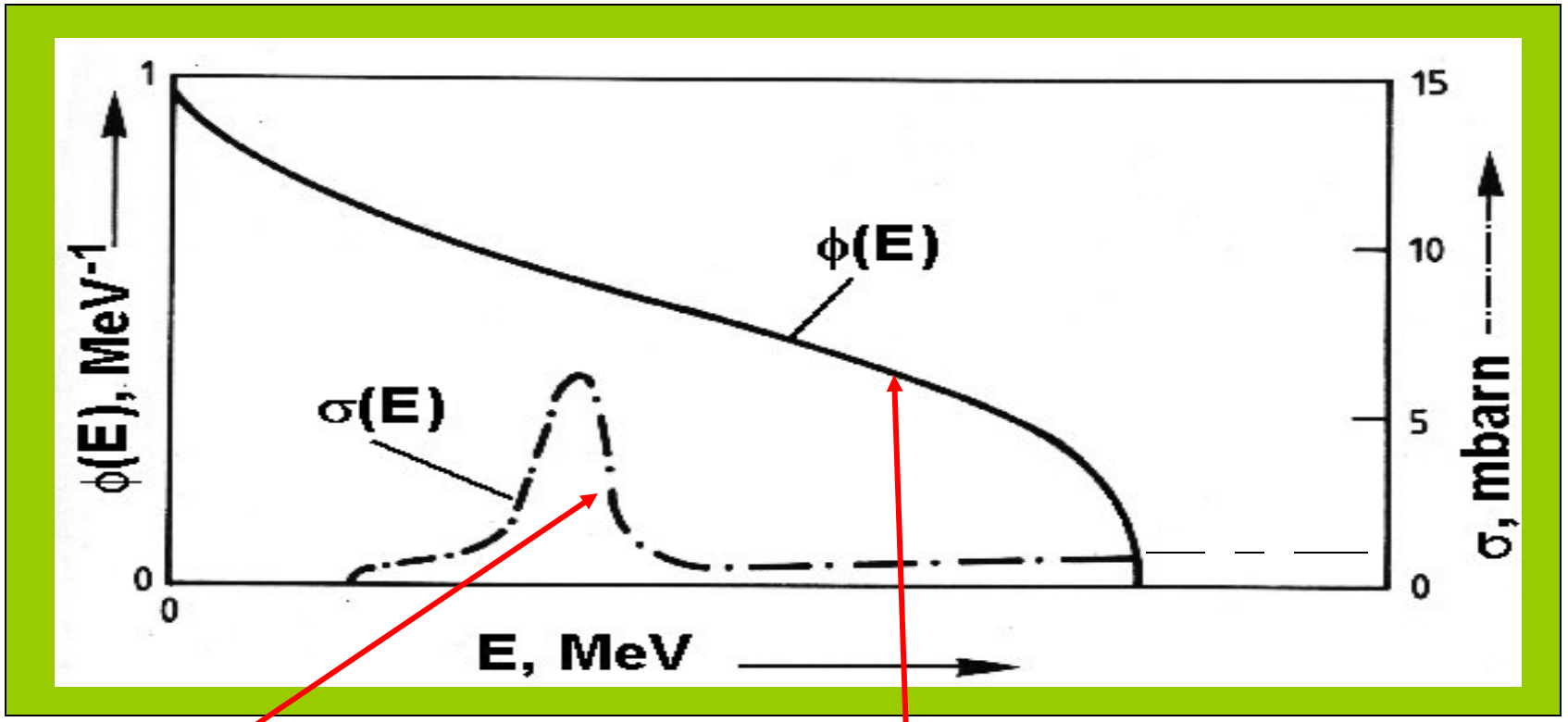


accelerators ← → reactors

Yields are high!!!:

a fact not often appreciated:

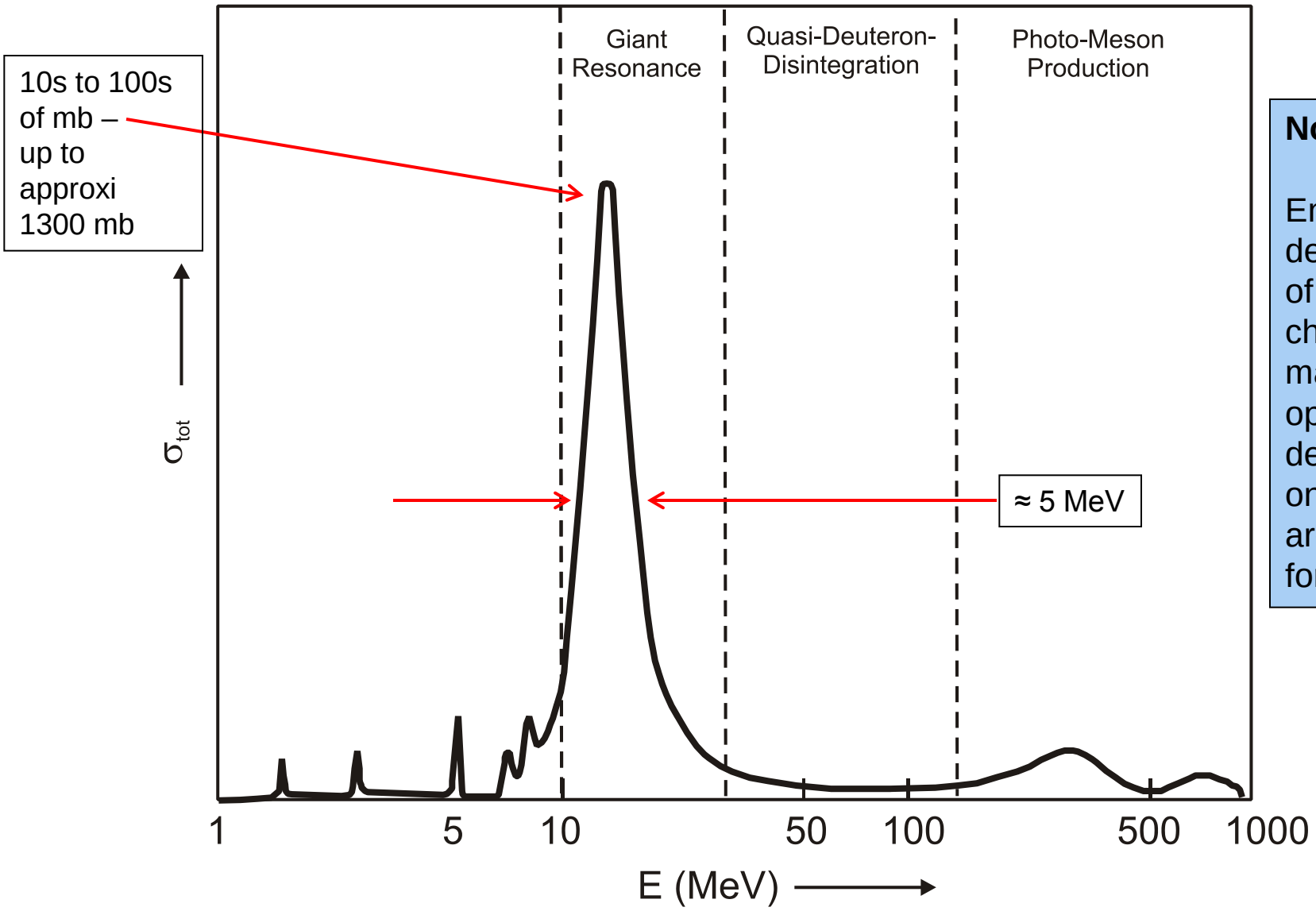
$$Y \propto M \int_{E_S}^{E_{\max}} \varphi(E_\gamma) \cdot \sigma(E_\gamma)\, dE_\gamma$$



σ(E): Cross-section of the nuclear reaction under study, from 10s mb ranging up to ≈ 1 b, with a width of approximately 5 MeV.

Φ(E): Flux density of the activating particles, e.g. bremsstrahlung photons produced by the electron accelerator.

Photo-Nuclear Cross Sections are NOT always small!



Note:

Energy dependence of reaction channels makes optimization dependent on what you are looking for.

Modest photo-nuclear peak cross sections are compensated by thick targets, a broad resonance cross section, and integration over a broad energy spectrum

$$Y \propto M \int_{E_S}^{E_{\max}} \varphi(E_\gamma) \cdot \sigma(E_\gamma)\, dE_\gamma$$

Rough Needs:

E_e ≥ roughly 30 - 50 MeV, for MOST reaction channels
P > few kW, up to hundreds of kW
Bremsstrahlung Target ≈ 1 Radiation Length,
Sample ≈ typically g/cm^2, but one can go considerably thicker or thinner

Note:

Energy dependence of reaction channels makes optimization dependent on what you are trying to produce.

$$Y \propto M \int_{E_S}^{E_{\max}} \varphi(E_\gamma) \cdot \sigma(E_\gamma)\, dE_\gamma$$

Figure of Merit:

For some cases, notably (gamma, gamma'), one may optimize the statistical figure of merit (FOM) by using an electron beam energy below particle emission threshold. But even for reaction channels above particle emission threshold, there are statistical FOM tradeoffs to be made in choosing the electron beam energy.

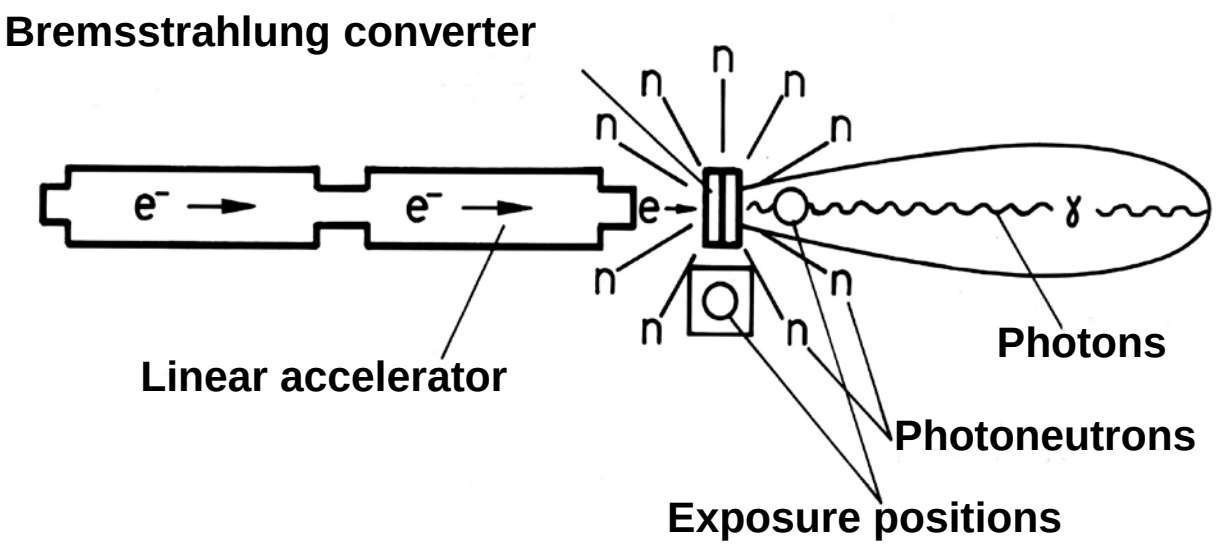
Isotope	Crude yield per 100 kW per week from 100g targets at 50 MeV electron beam energy
F-18*	9 kCi/wk
Cu-64*	10 kCi/wk
Ba-131*	15 kCi/wk
Mo-99*	2 kCi/wk (approx. 10% national need)
In-111*	2 kCi/wk
Y-88*	6 Ci/wk
Cu-67*	12 Ci/wk

These rates of production are very significant when compared to the national demand.

* - Taken from measured yield at 2 kW using IAC linac and scaled up by power, mass, beam energy, isotopic enrichment, geometric corrections, etc. Note that these rates are consistent with published rates of Segebade et al. and the Mo-99 estimates of TRIUMF.

Where does high power and CW fit in?

Production is done in a bremsstrahlung beam from an electron linac – typically need tens of kW of electron beam power to (if large scale production) hundreds of kW, at ≈ 30 - 100 MeV.



Two options: CW or conventional copper cavity linacs

For small scale production or regional production, namely for research isotopes and/or research into isotope production techniques, copper cavities at 10s of kW works well.

For production on a national scale, however, CW's **high power** is needed because one must go to 100s of kW, or higher.

So why hasn't photo-nuclear production been exploited?

In two words:

Specific Activity (SA)

(or lack thereof)

This is the Achilles Heel of photo-nuclear methods.

So why hasn't Photon Activation been exploited: Kinematic Recoil Methods….

Example: ^{99}Mo – photo-production via (gamma, n):

The maximum Specific Activity of ^{99}Mo in a post-gamma-irradiated pure ^{100}Mo target is in the 1-10 Ci/g range (see Bennett, 1998 or TRIUMF, 2008).

But if one separates the ^{99}Mo from a post-irradiated pure ^{100}Mo target, the answer is very different. Advances in materials science may enable one to produce:

better than 10 kCi/g specific activity of ^{99}Mo

on a laboratory scale (tens of uCi) – can it be scaled up?

So why hasn't Photon Activation been exploited? An alternative to kinematic recoil separation are chemical separations of (gamma, charge-particle) reactions….

Example: ^{67}Cu – photo-production via (gamma, p):

The maximum Specific Activity of ^{67}Cu in a post-gamma-irradiated ^{68}Zn target is dominated by the impurities in the target and the impurities transmitted in the separation processes.

Need 1-10 kCi/g SA (reliably) at roughly 100 mCi/dose for clinical trials … eventual routine use in medicine will probably require approx. 10 kCi/g SA.

(ANL, RPI and others have led this effort, in parallel with efforts by BNL, LANL, Trace Life Sciences, etc. to use light-ion reactions)

However, most have an interest in First Order ADS Simple Example of Power Multiplication (with an electron linac)

- If k=.95 and a 100 MeV electron accelerator (N = 6x10e12 n/s/kWb; kWb is electron beam power) the number of fissions is related to a kW of input beam power as:

  $N_f = 6.4 \times 10^{13}$ fiss/s/kWb.

- The fission power is: $P_{fiss} = 1.9$ kW/kWb (i.e. a gain of 1.9)

- If one operated at k=0.98, instead of k=0.95, the fission power is:

  $P_{fiss} = 4.9$ kW/kWb (i.e. a gain of 4.9)

Second Order (2 coupled multiplying assemblies) ADS with an electron linac:

Two multiplying assembles (A1 and A2) coupled, a fraction (f) of the fast neutrons created in the first assembly allowed to serve as the neutron source for the second one. The total number of fission neutrons created in assembly 2 is then: N2- fN/(1-k1), so that the number of fissions in assembly 2 is:

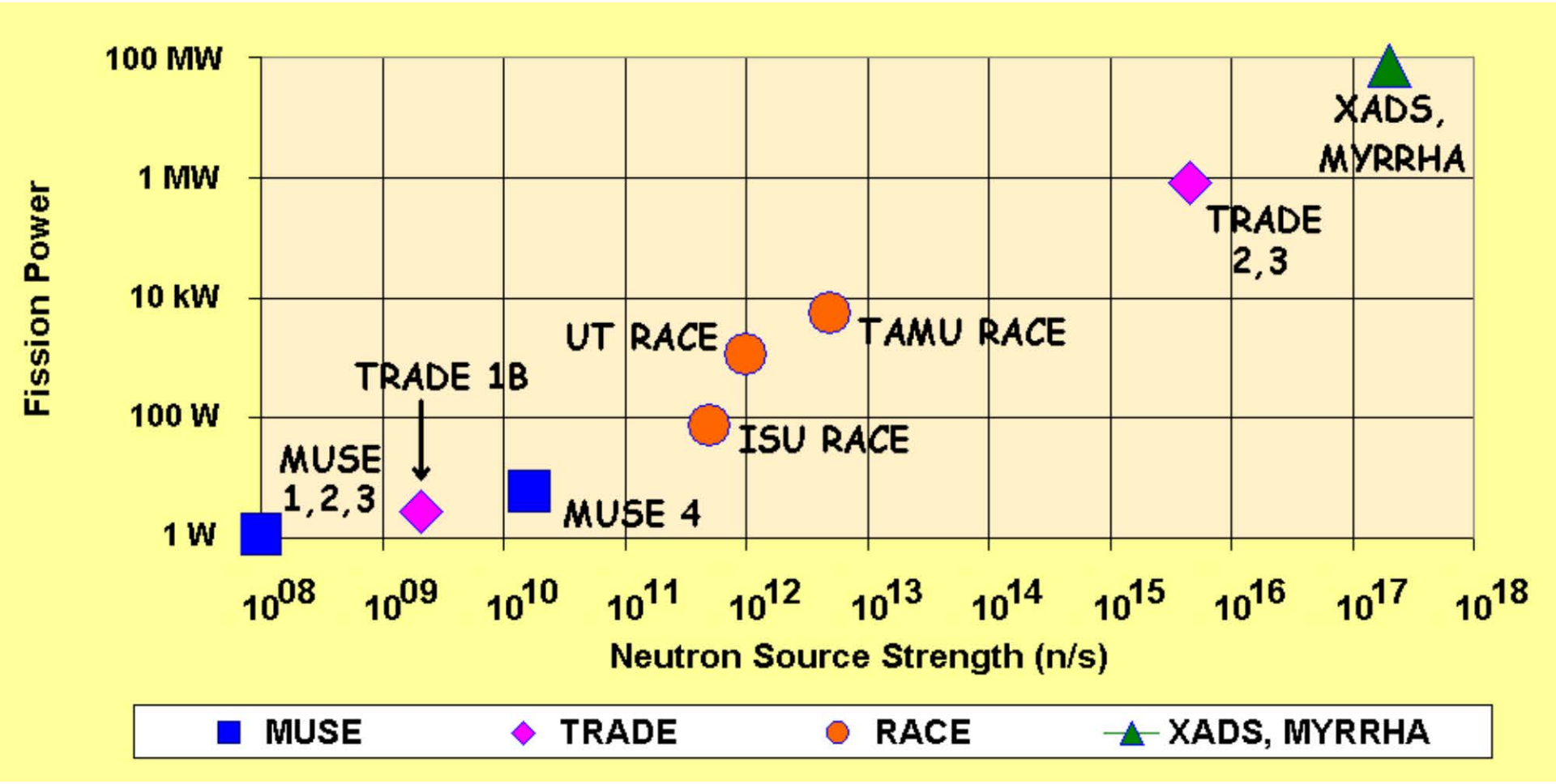
$$N_2 = fNk_2/v(1-k_1)(1-k_2)$$

If f = 1/2 and $k_1 = k_2 = 0.95$, fission power in the second assembly will be:

P_2= 9.9 kW/kWb (i.e. a gain of 9.9)

And if k1 = 0.98, k2 = 0.95, then: P_2= 25 kW/kWb (i.e. – a gain of 25)

Source strength and reactor/fission power comparisons:

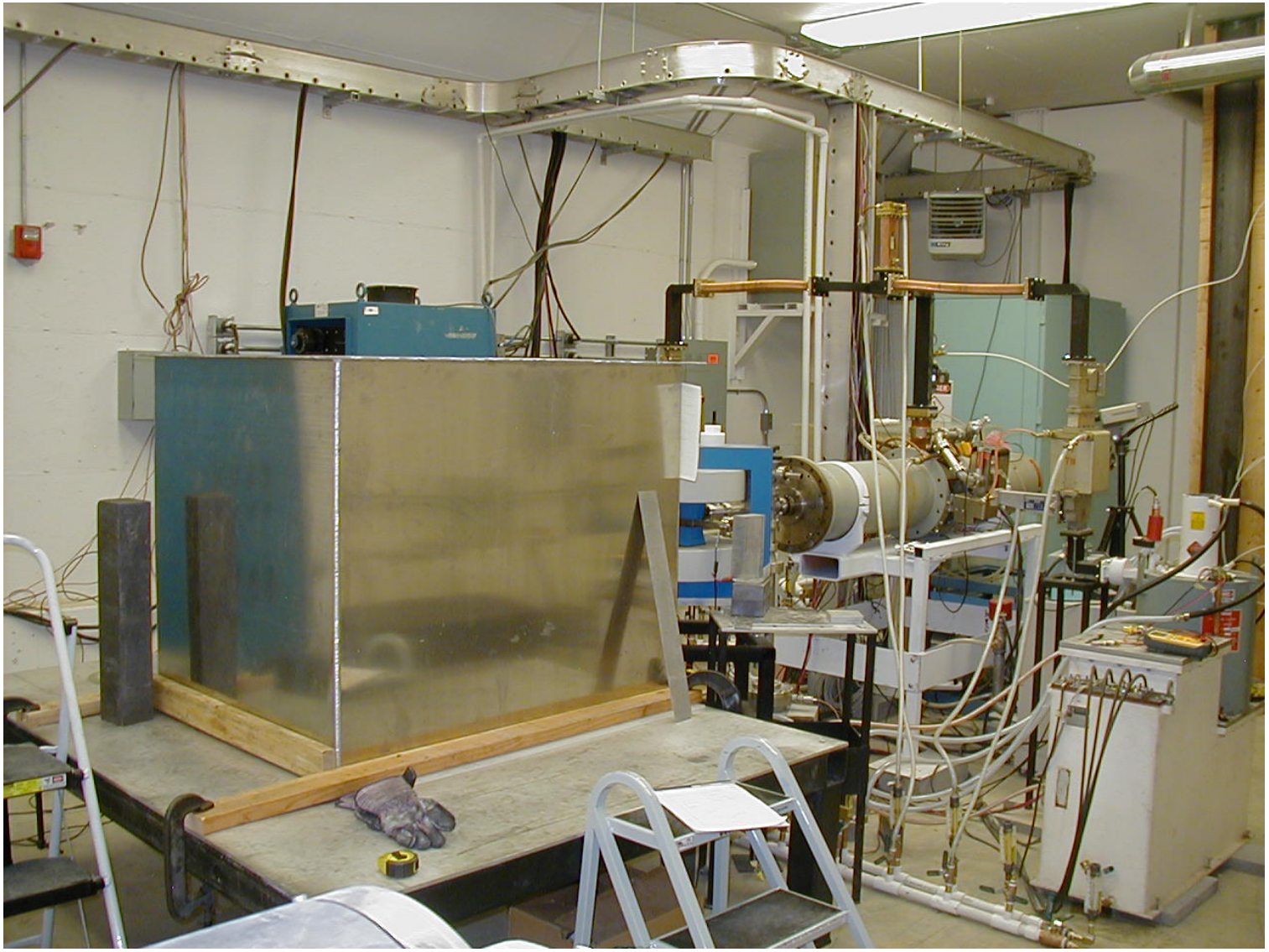


U.S. Reactor-Accelerator Coupling Experiment (RACE)

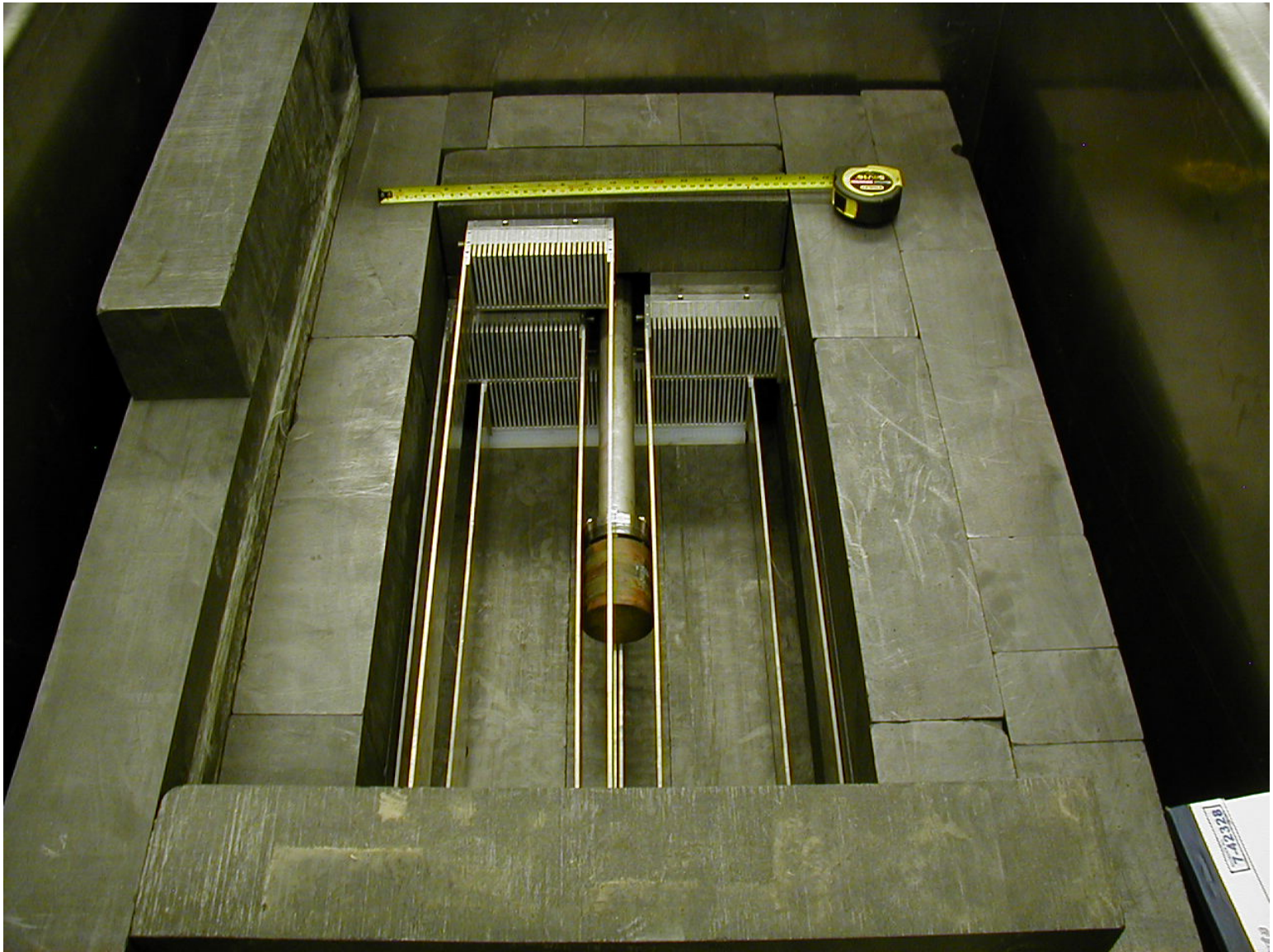
Idaho Accelerator Center

A 10 kW, 40 MeV electron linac for high-power applications can be very compact:

Original ISU RACE project:
ISU Accelerator and Sub-Critical Assembly:



Original ISU RACE fuel trays and target inside the graphite reflector



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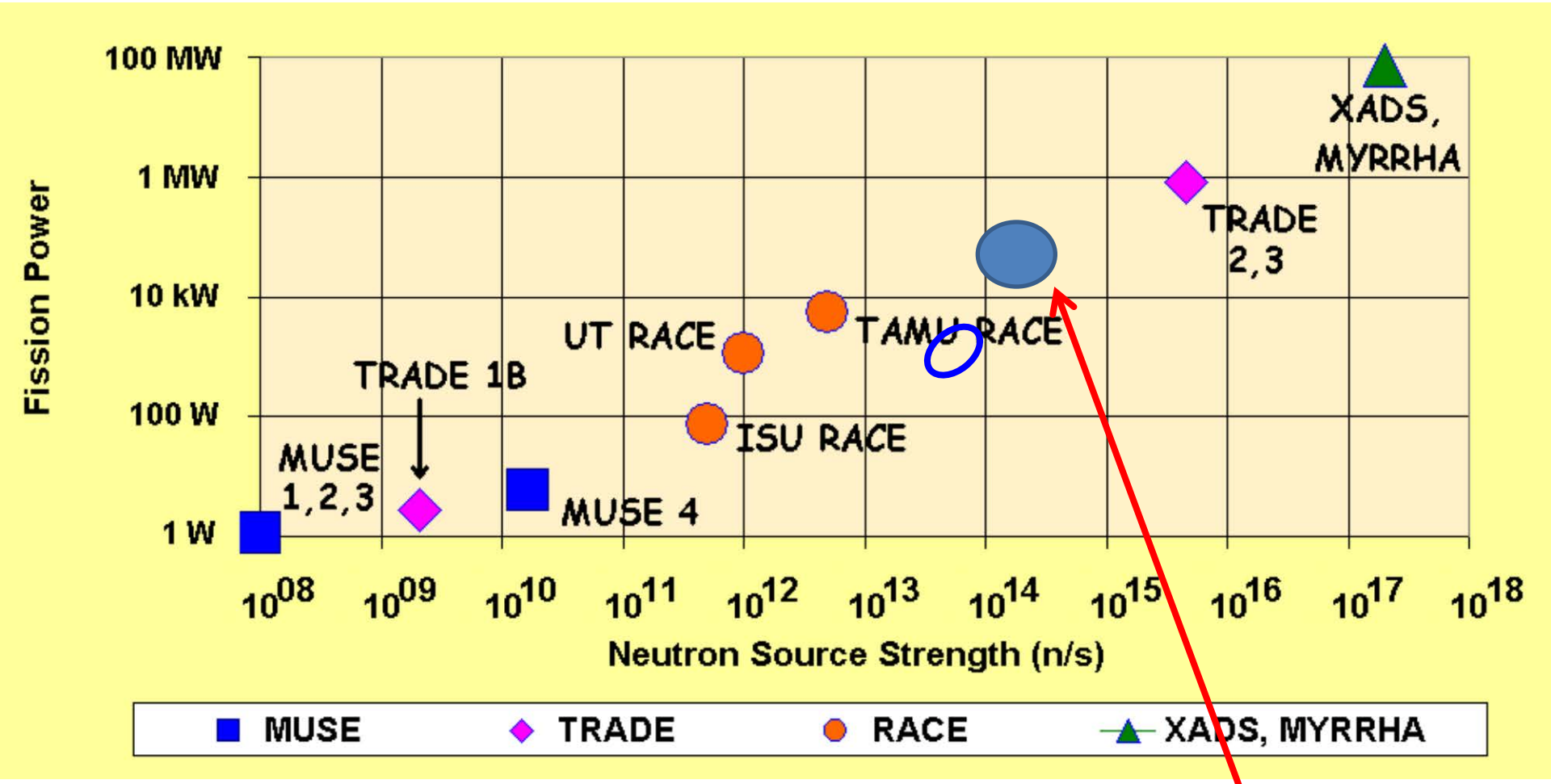

Follow-on work to U.S. Reactor-Accelerator Coupling Experiment is possible: (RACE, 2003 – 2006) was an ISU-led, DOE-funded research project (with UT, TAMU, UNLV, U. Mich., INL, ANL, LANL, CEA, ECATS)

- Design, model, and conduct ADS experiments
- Predict and analyze subcritical multiplication and source-driven transients
- Map source importance & flux
- Study ADS startup and shutdown
- Study dynamic effects of power at different sub-criticality levels (feedback vs. source effects)
- Study start-up/shut-down scenarios
- In general, study all relevant aspects of current/power/importance/control rod relations

Conclusions and Questions

- The speed of advancement and private/governmental support of ADS systems may hinge on how soon practical applications are possible.

- Certain practical applications, such as isotope production and fuel-cycle materials irradiation are practical NOW.

- Should there be more effort placed in these areas?

Thank You!