Towards Integral Electron Measurements of $^{233,235}U$ and $^{239,241}Pu$ at HFIR Decay Station

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On behalf of the working group

(ORNL-ANU-BNL- UTK)

WoNDRAM, June 2021

ORNL-BNL-Australian NU- UTK working group proposal

MOTIVATION:

35+ years old integral electron spectra emitted during the neutron-induced fission of nuclear fuel components, "Schreckenbach data" (ILL Grenoble), are still serving as an input to the reactor anti-neutrino flux analysis including the "reactor anti-neutrino anomaly" and its consequences for fundamental physics.
New results obtained by Kopeikin et al at the Kurchatov Institute (KI Moscow) do not agree with the ILL data.

The reactor anti-neutrino flux must be determined better than 1%, to solve the "reactor anti-neutrino anomaly" and to serve new generation of neutrino oscillations experiments near and far from nuclear reactors.

SOLUTION:

High Flux Isotope Reactor (HFIR) with the adjacent Radioisotope Engineering Development Center (REDC) at ORNL is the perfect place for the re-examination/measurement of integral electron spectra in fission products providing a suitable spectroscopy setup including electron spectrometer served by a neutron beam is implemented.

PROPOSAL:

Made unambiguous direct measurement of integral electron spectrum from n + ^{235}U,^{239}Pu and ^{241}Pu using unique HFIR/REDC resources and new suitable spectroscopy setup.

Once the experimental setup at HFIR is commissioned, the measurements can be extended to the spectroscopy of other actinides like ^{233}U to help reactor science and safeguards/non-proliferation studies. Integral γ-decay heat measurements (decay heat) can be refined, too.

“Schreckenbach data” 1981-1985 ILL Grenoble

The integral beta energy spectra of fission products were measured using a huge magnetic **spectrometer BILL,** about **14 meters** from the **~ 150 μg/cm^2**,18 cm^2 ^{235}U target (later **1 mg/cm^2**) sandwiched between two 7 mg/cm^2 Ni layers. Targets were irradiated for **15-30 hours**, with a neutron flux of **~ 10^{14} n/cm^2/s. Detection solid angle was very small, about 3x10^{-6} of 4 Pi.**

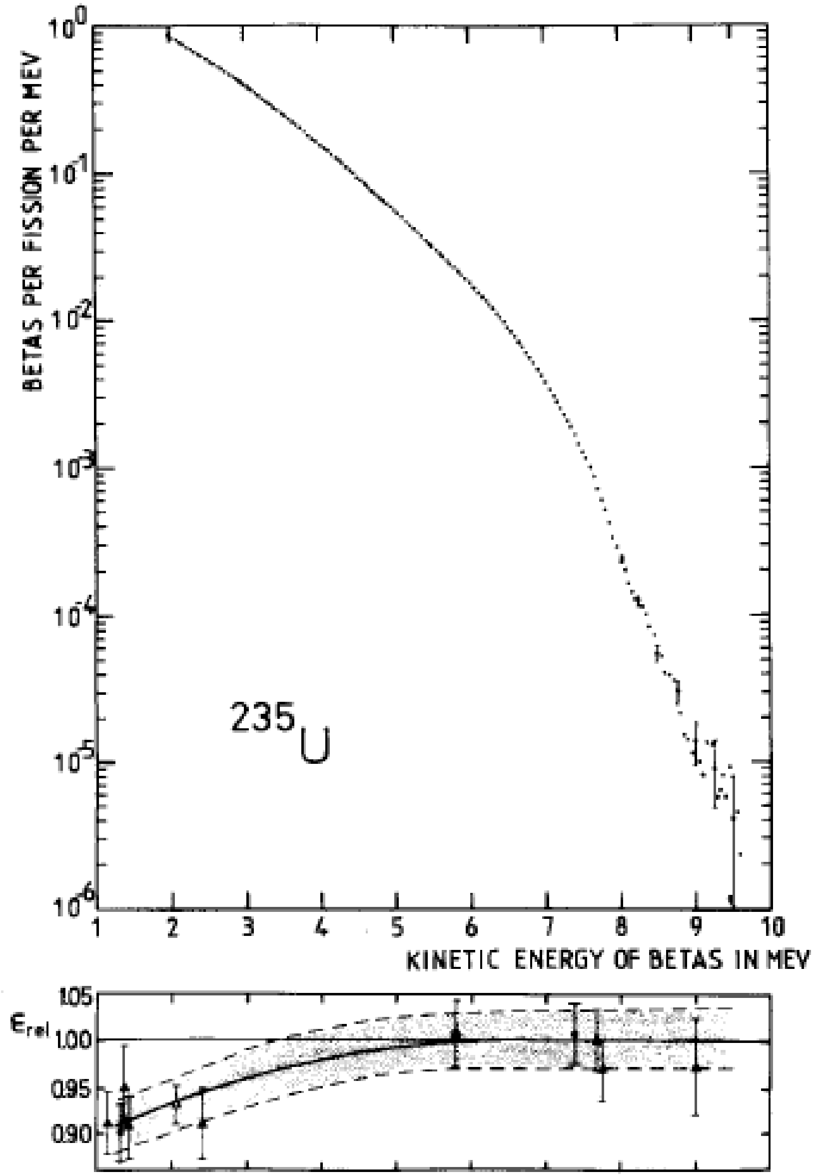
Electron energy was measured using a multi-wire ionization chamber in a transmission mode, with **good energy resolution** about ~ 10 keV at 3 MeV electrons, 50 keV bins, from ~ 1 to 10.5 MeV, 30 s to 150 s counting per bin.

Huge neutron flux, thin targets, but with a relatively thick backing, very large spectrometer with good energy resolution but very low transmission (efficiency), lower background due to the large distance from the target to electron detector. Measurements lasting ~ days after the method was established/verified.

Schreckenbach, Colvin, Gelletly,von Feilitzsch, Phys. Lett., 160B, 325, 1985

“Schreckenbach data”: integral electron energy spectra for n+^{235}U, ^{239}Pu and ^{241}Pu

Integral electron energy spectrum: over 6 orders in magnitude between 2 MeV and 9.5 MeV



BILL efficiency

The electron spectrum was deconvoluted into 25 components, of different end-point energies, assuming an “effective” atomic number Z=46 for fission products. Method was tested by using two effective atomic numbers, Z =36 and Z=55, with somewhat arbitrary weights between 2/3 and 1/3.

^{235}U integral anti-neutrino spectrum was deduced out of these 25 beta components and claimed to have a conversion error ~ 3% and total error of ~ 6.5% in 2-7.5 MeV range. **Small deficit of anti-neutrinos was within error bars, 97.5+-2.5%.**

In 2011, independent advanced analysis of these spectra (Huber 2011, Mueller 2011) led to the conclusions that the deficit is larger, ~ 95 (2)%, and reactor anti-neutrino anomaly was born.

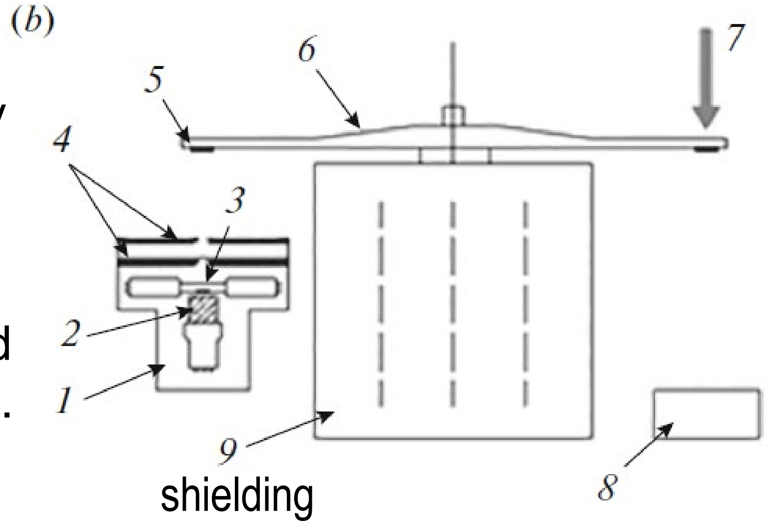
“Kopeikin data” ~ 2014-2021 Kurchatov Institute Moscow

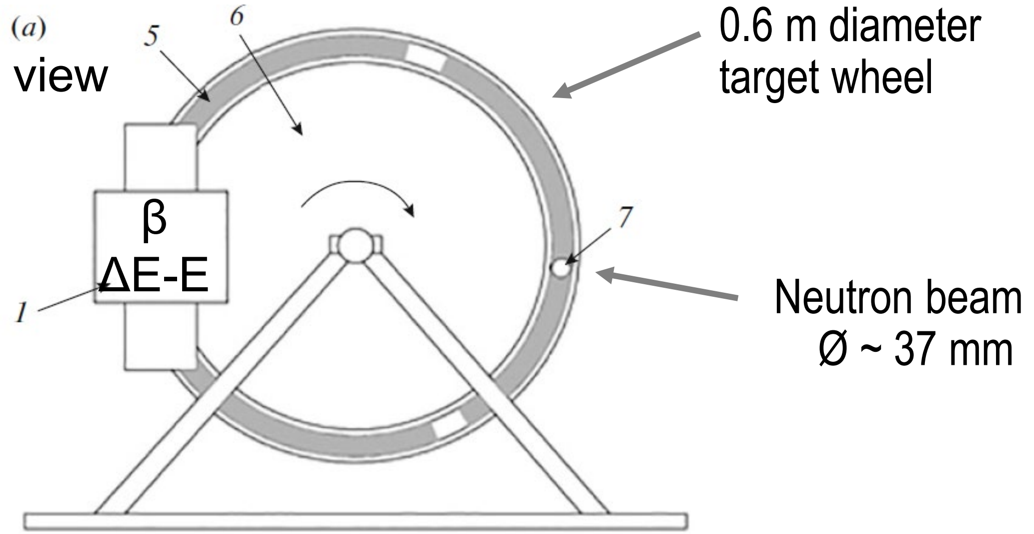
Kopeikin, Panin, Sabelnikov, Phys. At. Nuclei, 2021, 84, 1 and Balygin et al, Instr. Exp.Techniques, 2014, vol.57, no.1, 22
see also Borovoi et al, Yad. Fizika, 32, 1980, 1203

Around 2010-21, new measurements of integral electron flux from n+^{235}U/^{239}Pu were performed at the Kurchatov Institute (**KI**) Moscow, at the IR-8 reactor. Neutron flux up to **7x10^{6}** n/cm^{2}/s
Targets of ^{235}U and ^{239}Pu were 2x3 cm, **39 mg/cm^{2} thick**,16 targets each, at the rotating 60 cm diameter wheel, with **13 μg/cm$_{2}$** backing. 16 “background targets” (48 total), measurements in series of **6 hours**.

Top view
ΔE-E β-energy

fwhm 120 keV
at 1 MeV,
γ-bckg reduced
by a factor 200.





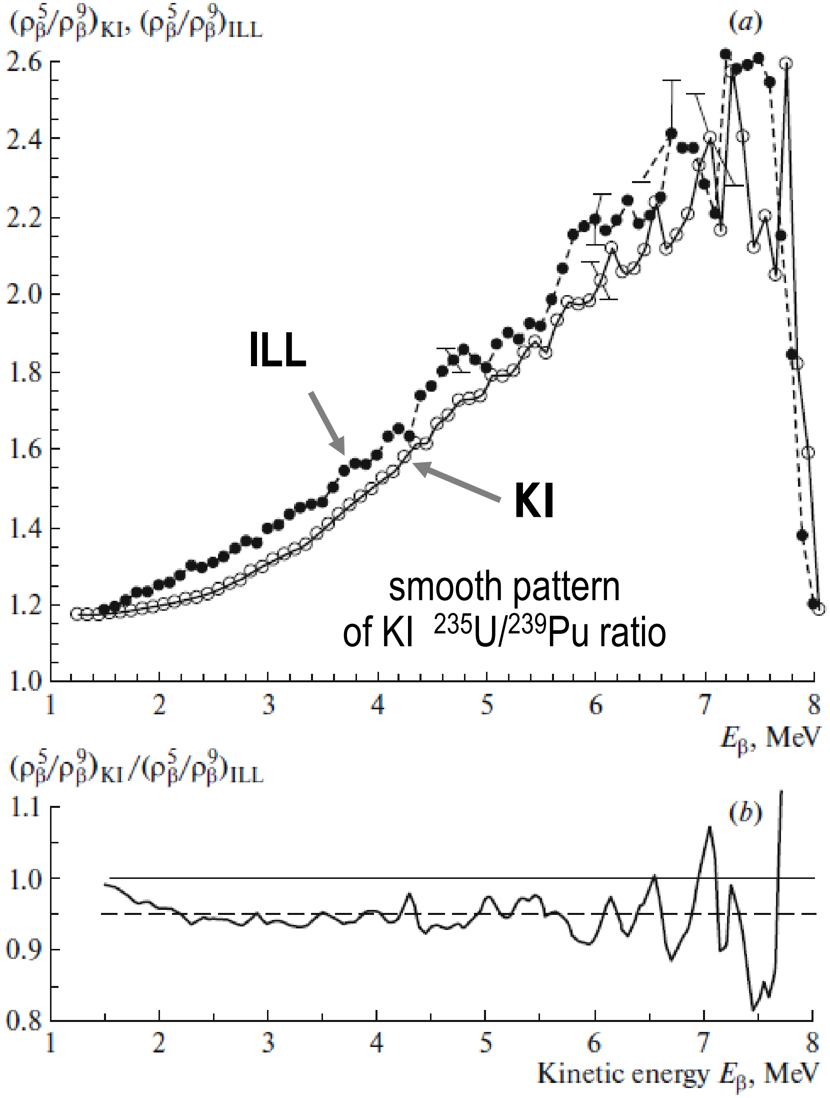
Moderate neutron flux at KI (HFIR 3*10^{7} n/cm^{2}/s), thick targets (!!??), small size setup with poor energy resolution but reasonable transmission (efficiency). Measurements lasting ~ days after the method was established, verified and calibrated.

Kopeikin's presentation of results: ratios of $^{235}U/^{239}Pu$ electron spectra

Ratio of ratios for electrons from $^{235}U/^{239}Pu$ fission

KI / ILL

~ 5% difference comparable with "reactor anomaly"



Large error bars at the high energy region 6 MeV – 8 MeV in both ILL and KI measurements

Fig. 3. Ratio of the cumulative spectra of beta particles from ^{235}U and ^{239}Pu fission products according to measurements at Institute Laue–Langevin (ILL) and Kurchatov Institute (KI): (*a*) ILL $^{235}U/^{239}Pu$ ratio of beta-particle spectra, $(\rho_\beta^5/\rho_\beta^9)_{ILL}$ [5, 6, 18] (closed circles connected by the dashed curve), and KI $^{235}U/^{239}Pu$ ratio of beta-particle spectra, $(\rho_\beta^5/\rho_\beta^9)_{KI}$, according to our present measurements (open circles connected by the solid curve); (*b*) comparison of the spectra from the present experiment, $(\rho_\beta^5/\rho_\beta^9)_{KI}$ and the experiment at ILL, $(\rho_\beta^5/\rho_\beta^9)_{ILL}$.

New measurement of integral electron spectra from neutron-induced fission of main reactor fuel components

ORNL's High Flux Isotope Reactor equipped with Decay Spectroscopy Station

ORNL: Rykaczewski, Allmond, Rasco, Crow, Cao **BNL:** Sonzogni, **ANU:** Kibedi, Stuchbery, Lane, **UTK**: Grzywacz

New HFIR based measurements following Kopeikin's compact setup should include:

(a) at least 10^7 n/cm2/s neutron flux, one HFIR cycle per year, proper fission rate normalization
(b) optimized targets with thin backing (stop fission fragments, let electrons out with minimal energy losses)
(c) two e-spectrometers, for monitoring at a fixed energy (~3 MeV), and a larger one scanning the electron energies
(d) good energy resolution (~ ten keV) and transmission (~%), 25 keV bins for the electron spectra
(e) meaningful high statistics of electron spectra, up to 9 MeV
(d) gamma detectors for gamma-gamma coincidences and normalization, later TAS for the integral decay heat
(f) radiation background reduction/passive shielding

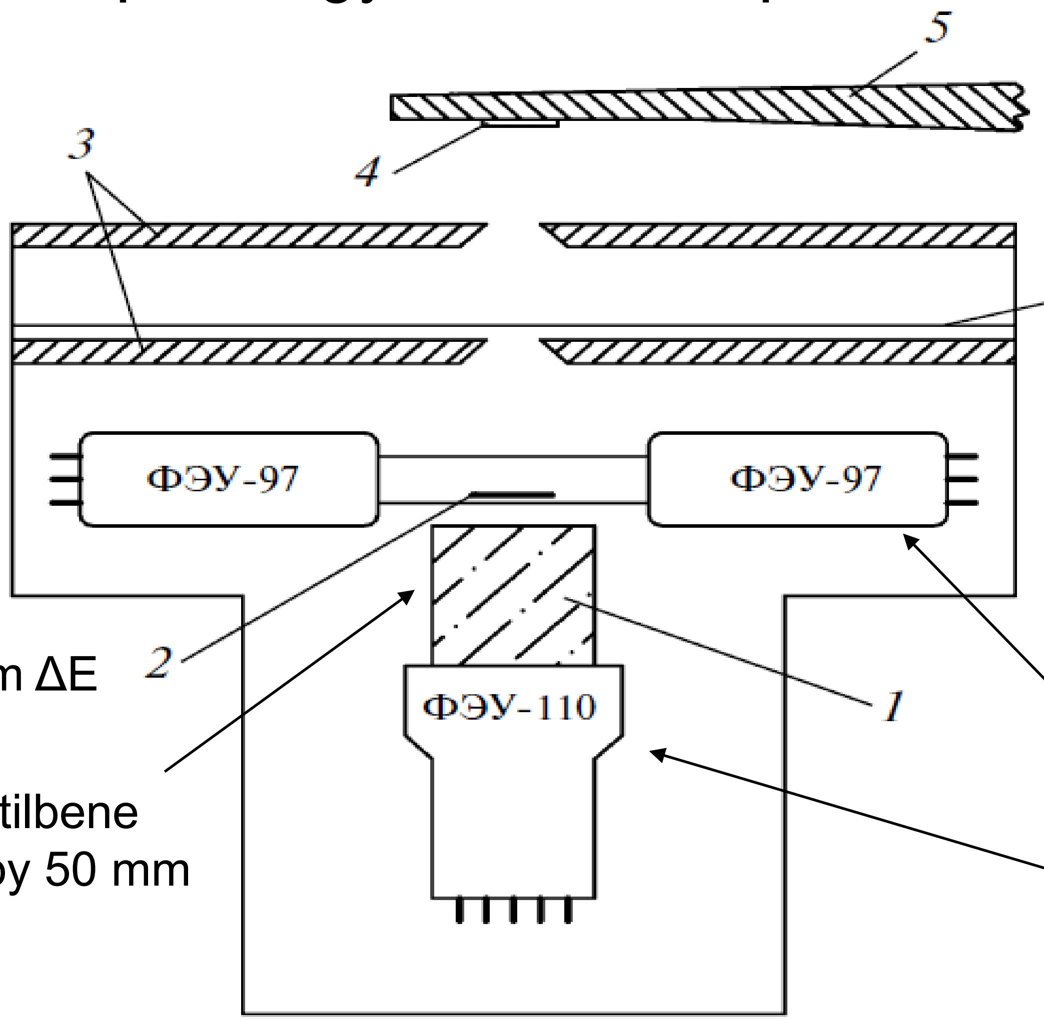
The measurement environment at HFIR, REDC made U/Pu targets and electron spectrometer are needed to start the experiments. We can initiate the measurements with few energy to verify some of ^{235}U/^{239}Pu electron spectra ratios, with respect to the ILL and KI results.

Total cost in ~ $15 M range, includes new equipment and MANPOWER

About ~6 years project, with one 25-days cycle/year at one of HFIR's beam lines.

ΔE-E β-energy detector, top view

Moderate neutron flux (HFIR 3*10^7 n/cm2/s),
thick targets (??), small size setup
with poor energy resolution
but reasonable transmission (efficiency).
Measurements lasting ~ days
after the method is established/verified.



0.16 mm ΔE

Stilbene
50 by 50 mm

1 MeV γ-background
reduction ~ 200

12% fwhm for
1 MeV electrons
120 keV !!
efficiency ~ few %

old PMTs

Fig. 2. Diagram of the β spectrometer: (*1*) main *E* detector, (*2*) drift Δ*E* detector, (*3*) diaphragms, (*4*) targets, (*5*) rotating disk-target holder, and (*6*) Mylar membrane.