Tale of Two (simple) Nuclides in ENSDF

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Presented by B. Singh

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Fate of some Nuclides in ENSDF

- ^{195}Os: 56-year old story in ENSDF about identification and half-life of ground state. Accepted from 1958 to 1994 or so; rejected in 1999. Recovery effort in progress now!
- ^{195}Ir: beta-daughter of ^{195}Os: 40-year old small but significant error in the evaluation of ground-state half-life in ENSDF.
- ^{53}Cu: this nuclide has been in ENSDF since 1999, with no questionable assignment as to its identification, but only in 2013, there is a publication based on experimental data about its being unbound, and the 2013 paper published as a result of my query to author.

^{195}Os: resurrection of a fallen nuclide!

^{195}Os (Z=76, N=119)

Identification and half-life of ground state ?

Information in databases:

ENSDF (2006): 0.0 ?, $T_{1/2}$ ~ 9 min (gross theory-1973): header comment: 1957Ba08 observed a 6.5-min activity …… 1974CoYN state that this activity is assigned to ^{81}Rb.

NDS (1999) : 0.0?: header comments list $T_{1/2}$ ~ 9 min and same as above for isotopic identification. In 1999-NDS, ^{195}Os is the most neutron rich in A=195.

NDS (1994): 0.0?, $T_{1/2}$ =6.5 min: cautionary comment from 1974CoYN.

NDS (1989,1984,1978): 0.0, $T_{1/2}$ =6.5 min: cautionary comment from 1974CoYN

NDS (1972, 1960-65): 0.0, $T_{1/2}$ =6.5 min

^{195}Os: resurrection of a fallen nuclide!

NUBASE (2012): $T_{1/2}$ =9 min (syst), year of discovery=2004. Comment: identification in 1957Ba08 with $T_{1/2}$ =6.5 m has been questioned, see ENSDF'07.

NUBASE(2003): $T_{1/2}$ =6.5 min: cautionary comment given; reference to 1999-NDS

NUBASE(1997): $T_{1/2}$ =6.5 min; not questioned

NUDAT 2.6: 0.0?, $T_{1/2}$ ~ 9 min : follows ENSDF and/or Wallet Cards

Wallet Cards (2011): $T_{1/2}$ ~ 9 m

TOI (1958, 1968, 1978, 1996), TORI (1986): all have 0.0, T1/2=6.5 min, not questionable. ^{195}Os first introduced in TOI-1958. In Seaborg's grading system A to G for isotopic assignment, ^{195}Os was assigned grade 'B' in TOI-1968, 1978.

Wall Charts (KAPL,GE) of Nuclides (2009, 2002): ^{195}Os ?, T1/2=6.5 m.

Wall Chart (GE) (1989 and earlier): ^{195}Os, T1/2=6.5 m: Not questionable.

Wall Chart (Karlsuhe) (2006): ^{195}Os, T1/2=6.5 m: Not questionable.

^{195}Os: discovery ?

- M. Thoennessen (MSU): isotope discovery project: ADNDT 98, 911 (2012): ^{195}Os discovered by J.J. Valiente-Dobón et al. at LBNL, PRC 69, 024316 (2004): multi-nucleon transfer reaction: 26-ns isomer found at 2230 keV in ^{195}Os. 1957Ba08, 1974CoYN, 1999-NDS were cited for rejection of 6.5-min activity assigned to ^{195}Os by
- Until about 2003, 1957Ba08 was the only experimental reference in NSR for ^{195}Os: Z. Naturforsch 12a, 520 (1957) (brief note of seven sentences!)

- Based on PRC paper from LBNL, 2004 was marked as year of discovery in 2012 ADNDT paper; as well as in NUBASE-12.
- This assignment raised some red flags!

1957Ba08: Z. Naturforsch. 12a, 520 (1957)

Ein neues Osmiumisotop von 6,5 min Halbwertszeit (Os^{195})

Von G. Baró und P. Rey

División Radioquímica, Comisión Nacional de la Energía Atómica, Buenos Aires

(Z. Naturforschg. 12 a, 520 [1957]; eingegangen am 20. Mai 1957)

Nach der Bestrahlung von Pt mit schnellen Neutronen der Reaktion Be(d 28 MeV, n) [1] wurde Os durch zweimalige Destillation als OsO_4 abgetrennt. In dieser Fraktion konnte eine Halbwertszeit von 6,5 min dem bisher noch unbekannten Os^{195} zugeordnet werden. Es wird durch den Prozeß $Pt^{198}(n, \alpha)Os^{195}$ gebildet. Seine mit Hilfe einer Feather-Analyse ermittelte β-Maximalenergie beträgt 2 MeV.

Die Identifizierung des neuen Os-Isotops ist einfach, da es durch Negatronenzerfall in das bekannte Ir^{195} von 2,2 Stunden Halbwertszeit [2] übergeht. Der genetische Zusammenhang wurde durch Intervalltrennungen von Ir aus der Os-Fraktion sichergestellt.

Die von Butement und Poe [2] für Ir^{195} und Os^{193} angegebenen Daten konnten bestätigt werden.

[1] Herrn S. Mayo und seinen Mitarbeitern danken wir für die Bestrahlungen mit dem Buenos Aires-Synchrozyklotron.

[2] F. D. S. Butement u. A. J. Poe, Phil. Mag. 45, 31 [1954].

^{195}Os: half-life ?

- June 21, 2013: Prof. Thoennessen sent an e-mail to Dr. Tuli:

  Dear Dr. Tuli,
  I have some comments regarding the current evaluation of 195Os. I think that the statement: "1957Ba08 observed a 6.5-min activity, in 198Pt(n,α) E=28 MeV, with Eβ(max)=2 MeV. 1957Ba08 assigned this activity to 195Os on the basis of genetic relationship to 195Ir. 1974CoYN state that the observed activity was later identified as due to 81Rb." is incorrect. Please let me know who is working on an update of the A = 195 mass chain and I would like to communicate my arguments why I think that this statement is not correct and 1957Ba08 should be credited with the correct half-life measurement.
  Thank you very much,
  Sincerely,
  Michael Thoennessen

^{195}Os problem

- Jagdish kindly sent me a copy of this email and his reply. I volunteered to look through this problem.
- Prof. Thoennessen mentioned that this issue was raised by Juan Flegenheimer in Argentina, who, at CNEA, was a colleague of authors of 1957Ba08 paper by Baro and Rey . Dr. Flegenheimer also brought to Michael Thoennessen's attention two detailed papers: P. Rey and G.B. Baro: Publ. Nac. Energ. Atom. Ser. Quim. **1**, 115-128 (1957); Proc. 2nd U.N. Int. Conf. on the Peaceful uses of Atomic Energy, Geneva (1958), vol. **14**, P/1570-1574 (1958).
- It took us a few weeks to get hold of the two papers. Paper in Spanish was kindly reviewed and partially translated by Eddie Browne.
- I obtained permission from NSDD group responsible for A=195 to re-evaluate ^{195}Os and its ground-state half-life for ENSDF.
- Michael Thoennessen, Juan Flegenheimer and I agreed to collaborate on the ENSDF evaluation of this nuclide.
- Step 1: get the above two papers entered in NSR with keywords. Incidentally, CNEA (Argentina) paper was cited by TOI-1968 and TOI-1978; but was never captured by ENSDF-NDS evaluators! U.N. Conf. paper has been missed all along, until now!

^{195}Os problem

- Step 2: reassess statement in 1974CoYN (Univ. Maryland Cyclotron Lab. rept. R. Colle, P. Gallagher and W.B. Walters, p119 (1974) : Decay of ^{195}Os and search for ^{196}Os

"Unfortunately, the then-existing assignment for ^{195}Ir has subsequently been identified as ^{81}Rb, arising from reactions induced in target impurities [2]. As a result, the present assignment of ^{195}Os will not withstand careful scrutiny". Assignment refers to Rey-Baro work of 1957; 74CoYN quoted both 1957Ba08 and their detailed CNEA paper.

Reference 2: 1968Ho01: NP-A 106, 382. This statement formed the basis of first questioning (from 1978-1994-NDS), then finally rejecting in 1999-NDS, the identification of ^{195}Os and its half-life of 6.5 min. ENSDF/NDS adopted property was copied in almost all the other databases, except fully in TOIs and Wall Charts!

^{195}Os: resolution of problem

- Step 3: We find that the time-line in the argument by 1974CoYN is incorrect. At the time of Rey-Baro work in 1957, accepted half-life of ^{195}Ir daughter was 140 min (2.3 h). But in 1962Cl07 (A.B. Claflin et al., NP 36, 652), half-life of ^{195}Ir was reported as 4.2 h by forming ^{195}Ir in (α,p) reaction on supposedly pure enriched ^{192}Os target. 1968Ho01 questioned the assignment by 1962Cl07 of an intense 190-keV γ ray to ^{195}Ir decay, and related this γ ray and 4.2(1) h half-life to ^{81}Rb formed by bromine impurity in target material used by 1962Cl07. 1968Ho01 made no mention of ^{195}Os or Rey-Baro work of 1957, who used (n,α) reaction on platinum and chemically separated osmium fraction. In Rey-Baro work, half-life of ^{195}Ir was also measured 2.2 h. Nevertheless, 1968Ho01 measured ground state half-life of 2.8(1) h and evidence for a 4.00(15) h isomer in ^{195}Ir; isomer later confirmed in other studies by 1968Ja06 + 1973Ja10

^{195}Os: resolution of problem

- Based on chemical separation of Os fraction, ^{195}Os – ^{195}Ir genetic relationship, and measured β-endpoint energy of 2 MeV (Q value is 2180(60) in AME-12), we consider assignment made by Rey-Baro in 1957 as firm, the same conclusion as reached in the evaluations of TOI-1968 and NDS-1960-65, 1972. Thus we recommend that identification of ^{195}Os ground state by Rey-Baro work of 1957 be reinstated. Now we move on to $T_{1/2}$ issue.
- Step 4: we decided to re-analyze composite decay curve presented by Rey-Baro using currently accepted half-lives of ^{195}Ir and ^{193}Os, the latter has been recently measured precisely as 29.830(18) h by Krane (PRC 85, 044319 (2012)). We find that half-life=2.5(2) h for ^{195}Ir ground state adopted in ENSDF/NDS, and also in almost all other databases since 1972-NDS is questionable. Thus we needed to re-evaluate ^{195}Ir g.s. half-life.

^{195}Ir g.s. $T_{1/2}$ problem

- 1952Ch18 (PR 86, 946); 1954Bu02 (Phil. Mag. 45, 31); 1961Ho10 (Jour. Phys. Soc. Japan 16, 841): $T_{1/2}$: 140 min (2.3 h) . But sources were mixed or decay curves not shown in papers to assess quality of data.
- 1962Cl07 (NP 36, 652): T1/2=4.2 h from decay curve of an intense 190-keV gamma.
- 1968Ho01 (NP-A 106, 382): questioned 1962Cl07 result, 190-keV gamma assigned to ^{81}Rb produced from Br impurity in sample (from same enriched material stock as used by 1962Cl07). They measured T1/2 as 2.8(1) h from decay curves of 99- and 129-keV gamma rays. They also identified an isomer in ^{195}Ir with T1/2=4.00(15) h. The ground-state transitions of 99- and 129-keV in ^{195}Pt were assigned only to the decay of 2.8-h activity.
- 1968Ja06 (NP-A 115, 321) + 1973Ja10 (Z. Physik 261, 95): confirmed the identification of isomer with T1/2=3.67(8) h, but measured the T1/2 of g.s. As 2.3(2) h from a decomposition of decay curve for 99-keV gamma ray which they assigned to both the g.s. and isomer decay.

^{195}Ir g.s. $T_{1/2}$ problem

- 1972-NDS evaluation: adopted 2.5(2) h from un-weighted average of 2.8(1) h from 1968Ho01 and 2.3(2) h from 1968Ja06. Since that time this life has been reported in various databases with no questions asked.
- Two issues with NDS adopted $T_{1/2}$:
- 1968Ho01 value is high because they erroneously assigned 99-keV gamma ray only to the decay of shorter-lived g.s. and did not account for admixture of longer-lived isomer. In our opinion, this value should have been discarded in the averaging procedure.
- 1957Rey-Baro work had also measured half-life of ^{195}Ir, which no data evaluator captured. After the decay of ^{195}Os from Os fraction, they chemically separated Iridium fraction and measured half-life of 2.2 h. This activity of Ir is expected to be purely from the ground state since it is populated by ^{195}Os g.s. decay; from systematics, spin of ^{195}Os g.s. is expected to be (3/2-) (see 2012Re19), and ^{195}Os isomer as (13/2+). Spins of ^{195}Ir g.s. and isomer are known to be 3/2+ and 11/2-, respectively

^{195}Ir g.s. $T_{1/2}$ problem

- We decided to re-fit the decay curves of Rey-Baro work of 1957. We tried to obtain data points from authors. Rey at 92 is alive but his current interest is cosmology not isotopes. Just received a word a couple of days back that original data points were not available.
- Digitized composite decay curve as well as 195Ir decay curve at three places: McMaster, MSU and XFOR code (this was done in India); this allowed us to assign uncertainties due to digitizing process in addition to statistical errors from Poisson statistics.
- Fitting of 195Ir decay curve gave a value 2.29(17) h, in excellent agreement with 2.3(2) h from 1968Ja06, and in disagreement with 2.8(1) h from 1968Ho01. Thus we recommend 2.29(17) h or 2.3(2) h for ENSDF, replacing the earlier value of 2.5(2)h and discarding 1968Ho01 result.
- Using 2.29(17) h for ^{195}Ir, 29.830(18) h for ^{193}Os, we obtain $T_{1/2}$=6.5(11) min for ^{195}Os g.s. Thus we will recommend this half-life in revised ENSDF evaluation. BACK TO ORIGINAL VALUE IN SEABORG's TOI (1958) TABLE!

^{195}Ir: 1957Re01 + 1958ReZZ

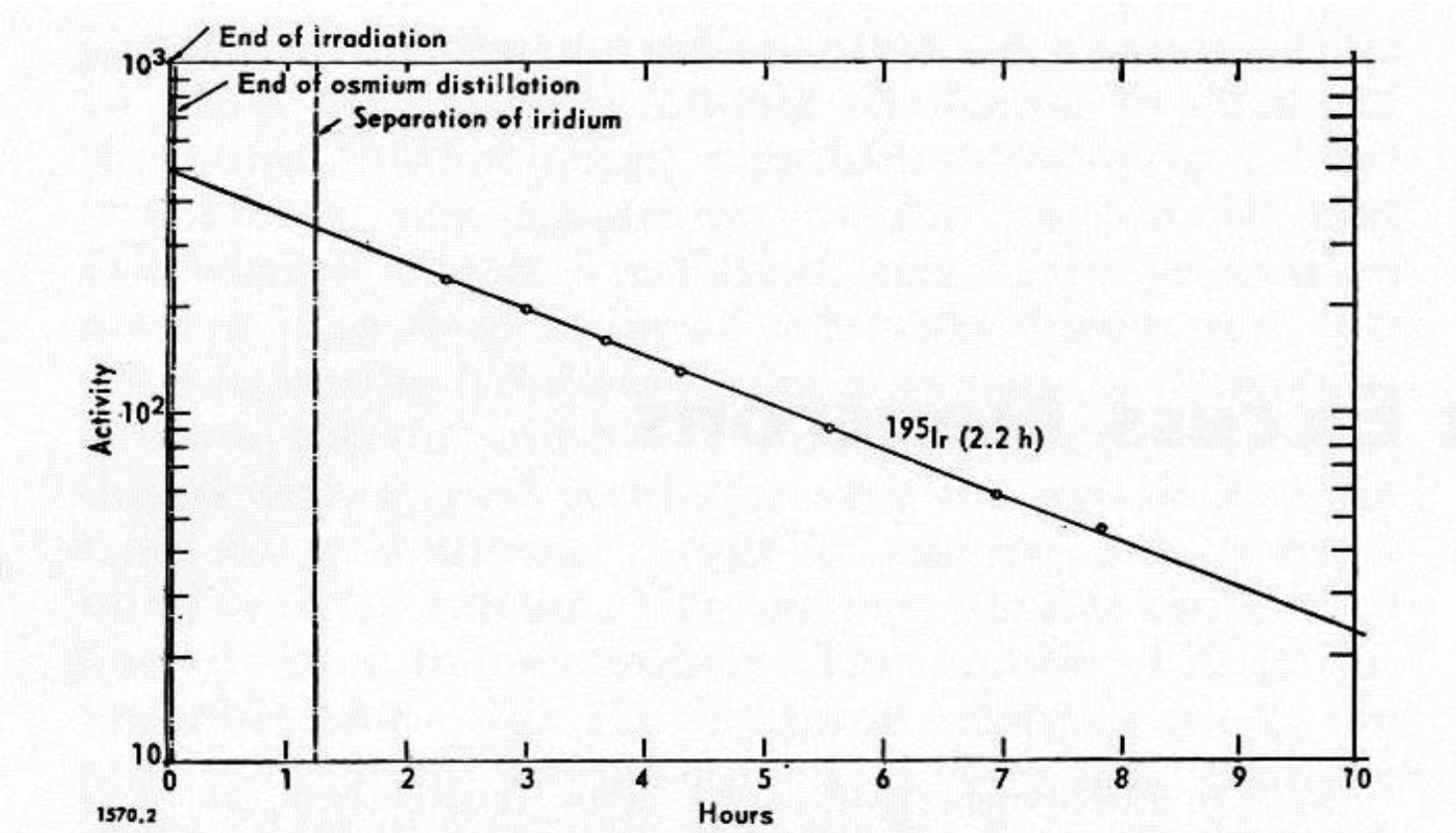


Figure 2. Disintegration curve for Ir^{195} separated from the osmium fraction after 50 min

^{195}Ir: 1957Re01 + 1958ReZZ

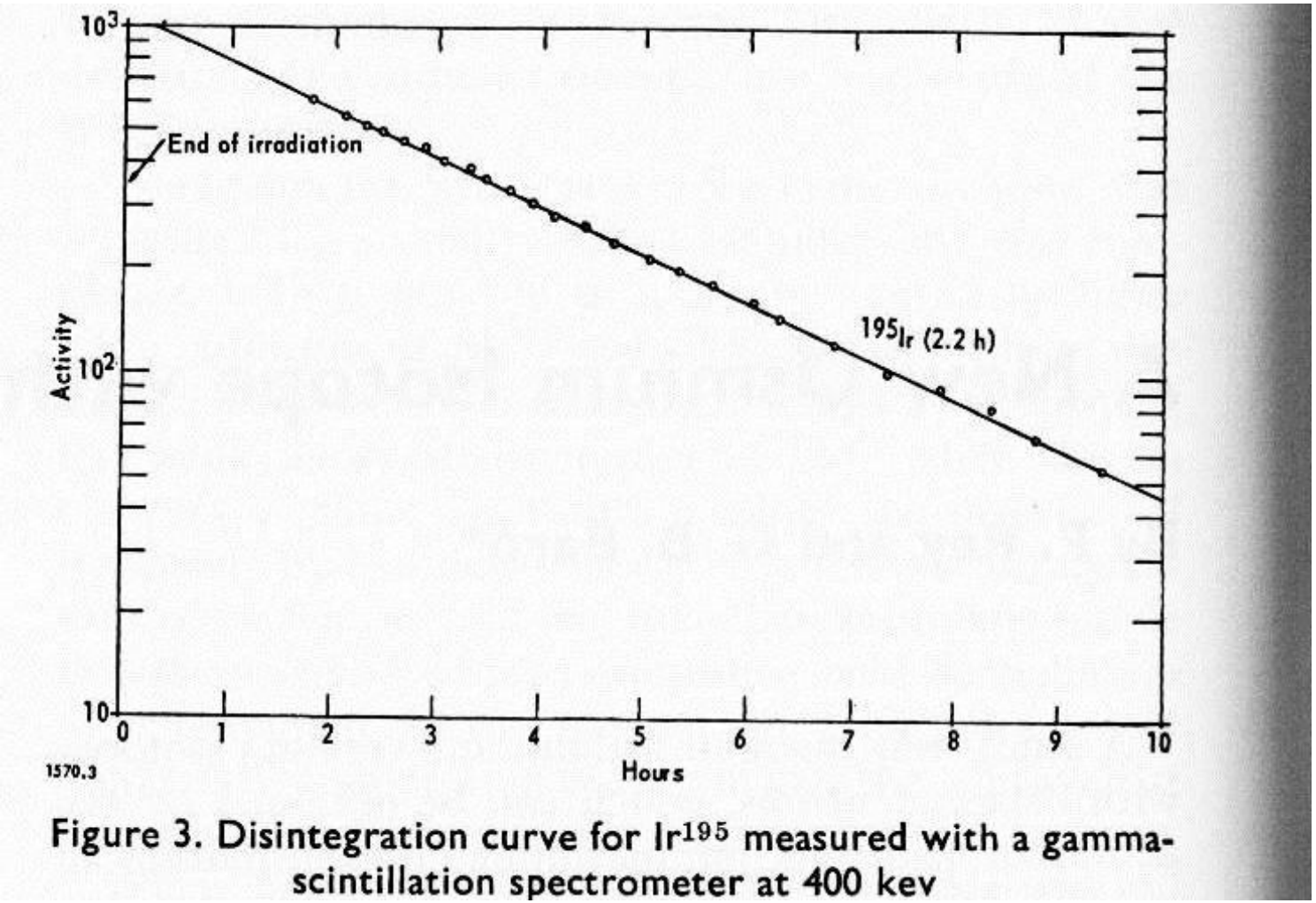


Figure 3. Disintegration curve for Ir^{195} measured with a gamma-scintillation spectrometer at 400 kev

^{195}Os: 1958ReZZ + 1957Re01

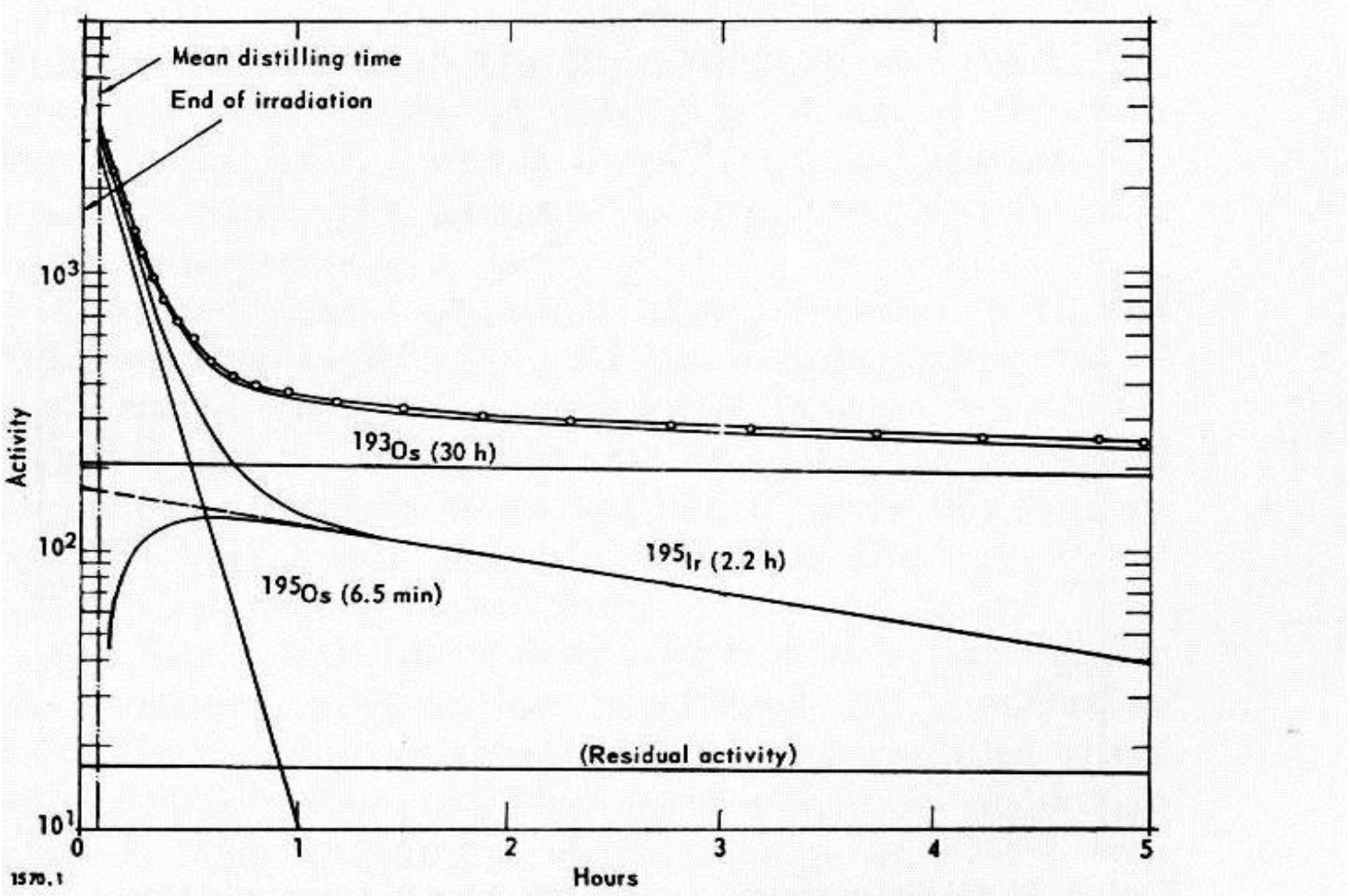
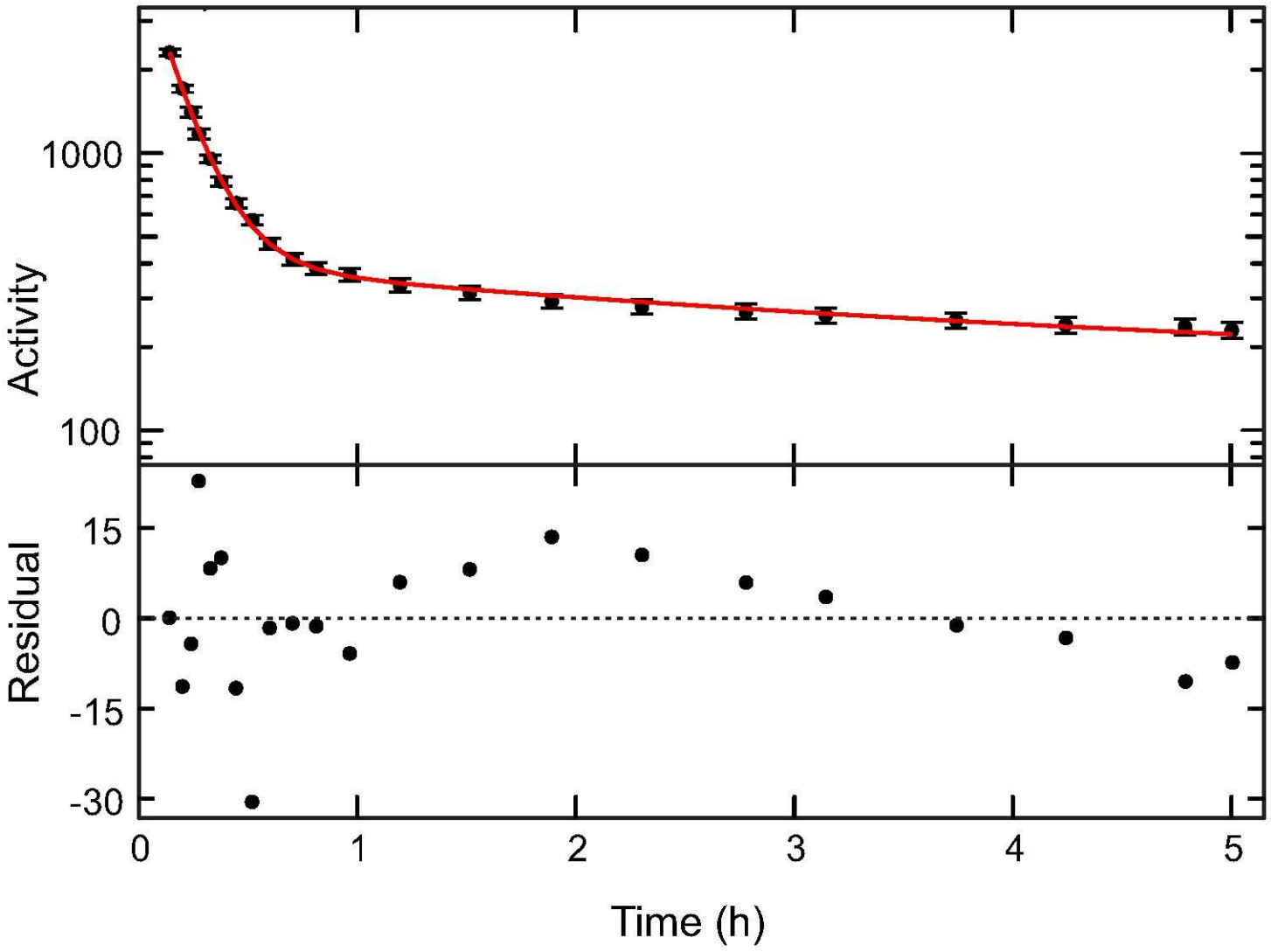
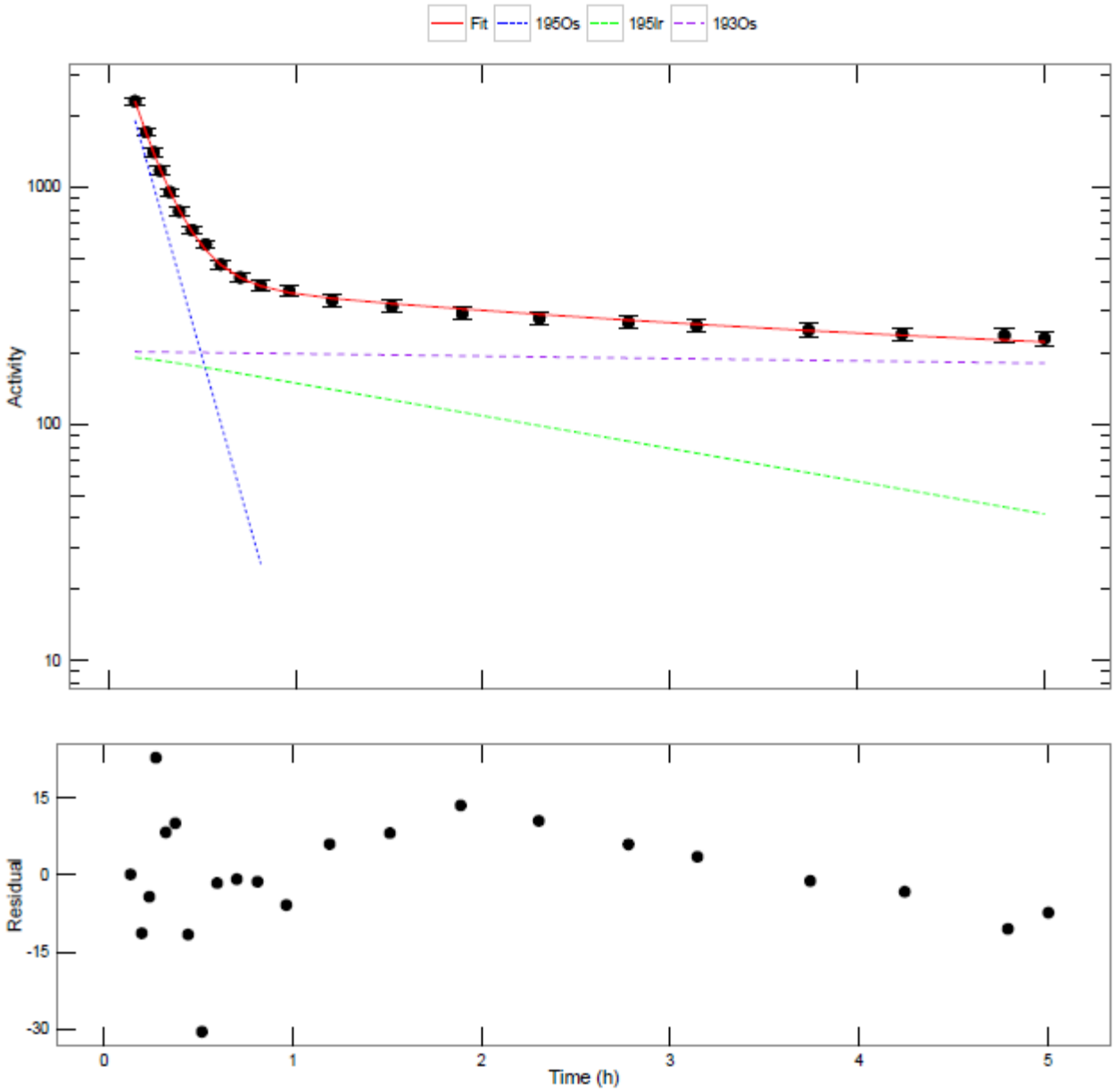


Figure 1. Disintegration curve for the osmium fraction separated from neutron-irradiated platinum

$^{195}Os + ^{195}Ir + ^{193}Os$




Fit
195Os
195Ir
193Os
Activity
1000
100
10
Residual
15
0
-15
-30
0
1
2
3
4
5
Time (h)

^{195}Os: up-to-date experiments

- 2004Va03: LBNL: PRC 69, 024316: 26-ns, high-spin isomer, at ~2.2 MeV; no information about g.s.

- 2005Ca02: GSI: EPJ-A 23, 201: 26-ns, high-spin isomer

- 2011St21: GSI: PRC 84, 044313: 34.0-ns, high-spin isomer

- 2012Re19: GSI: PRC 86, 054321: precise mass measurements of fully-ionized (bare) ^{195}Os ground state and an isomer at 454(12) keV with half-life of either 32(+154-16) min or 16 (+29-7) min. The ground state is expected to be (3/2-) and isomer (13/2+). I enquired the authors about g.s. $T_{1/2}$, but no response as yet.

^{195}Os, ^{195}Ir: draft of a brief paper

Revisiting the half-lives of ^{195}Os and ^{195}Ir

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Abstract: Currently the half-life of ^{195}Os is listed as unknown in most databases because the value of the only available measurement had been reassigned. We argue that the original assignment is correct and re-evaluate the half-life of ^{195}Os to be 6.5(11) min, consistent with the original measurement. We also recommend the half-life of ^{195}Ir as 2.29(17) h, instead of the currently accepted value of 2.5(2) h.

^{53}Cu (Z=29,N=24) nuclide in ENSDF

1999-NDS, 2009-NDS: isotopic identification from (1994BlZW,1997Au04); not questionable. Decay modes listed as %EC+%B+=?, %P=?. In 2009-NDS, reference 2003Au02 (NUBASE-2003) was added.

NUBASE-1997: first listed ^{53}Cu nuclide, ID from 1993Bl.A

NUBASE-2003, 2012: same as in NUBASE-1997; references to ENSDF-1999, 2009

GE Wall Chart-2009 does not list this nuclide

Karlsruhe Wall Chart-2006 lists this nuclide as identified.

- 1994BLZW (1993Bl.A in NUBASE-1997, 2003, 2012): GSI-94-1, p53: B. Blank et al., "……*In another measurement, the isotopes ^{53,54}Cu and ^{49}Co have been found to be unbound*. …."
- This report was followed up by 1994BL10: B. Blank et al., Phys. Rev. C50, 2398; with no mention of ^{53}Cu nuclide in this paper. There was also a later work from GANIL: 2005BL15: B. Blank et al., PRL 94, 232501; where no mention was made about ^{53}Cu nuclide.

^{53}Cu

Feb 18, 2013

Dear Dr. Blank:

I am writing in connection with status of Cu-53 nuclide for its lab production or identification or upper limit of cross section. In databases such as ENSDF and NUBASE-12, Cu-53 seems listed with some certainty, citing reference to your report in GSI-94-1, p53 (1994). However, your paper PRC 50, 2398 (1994) on the same topic as GSI report does not seem to mention Cu-53 nuclide, except in figure 6b, where it seems there may be ~2 counts associated with Cu-53. To my knowledge there is no other reference in literature about experimental work on this nuclide.
I would greatly appreciate your comments on the current status of Cu-53 nuclidic identification or not, so that it can be correctly/realistically presented in ENSDF database on Brookhaven lab's webpage: www.nndc.bnl.gov/ensdf/.

Thanks and best regards,
Balraj

^{53}Cu nuclide in ENSDF

Feb 18, 2013:

Dear Balraj Singh,

according to my knowledge there was no attempt to search for Cu53, simply because everybody believes that it is unbound. In our paper on the observation of Zn54 (PRL 94 (2005) 232501, attached) we could have done this, because we were optimized on Zn54 and Cu53 is a close neighbor. So I do it here quickly. I hope this helps.

The transmission of the LISE separator for Zn54 is 35%, for Cu53 it is calculated to be 45 %.

The production cross section of Zn54 can be estimated (by the EPAX formula and a factor of 10 loss per nucleon pick-up) to be 4e-13b, for Cu53 one obtains 1e-10 b. In figure 1 of the paper, one can clearly see 8 counts of Zn54. With the increased transmission (a minor factor) and the difference in production rate expected, it is evident that we should have observed hundreds of Cu53 which is evidently not the case. There might be one or two counts in the spectrum, but this is typically the background one would expect.

An "only experimental" evidence comes from the spectrum alone: Cu53 lays between Zn54 and Ni51 and as everything is expected to go smoothly, one expects the rates for Cu53 to be somewhere between Zn54 and Ni51 which clearly is not the case.

So, for me there is no doubt that Cu53 is unbound. All mass models give it unbound (the latest AME by about 1.9(10) MeV, the Garvey-Kelson relation by 1.6 MeV, the Antony-Pape relation by the same amount...

I hope this helps. If I can do more, please let me know...

Best regards

Bertram Blank

^{53}Cu

Feb 26, 2013

Dear Balraj Singh,
here is a first version of a paper on the instability of Cu53. Have look and let me know, whether this is what you need.
In particular, the remark with respect to NNDC could be modified....

Best regards
Bertram

^{53}Cu: new paper: 2013BL04

Particle instability of ^{53}Cu

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Abstract. The most proton-rich particle-stable or quasi-stable copper isotope identified in projectile fragmentation reactions is ^{55}Cu. ^{54}Cu was found unbound a few years ago in such an experiment. The present note reports on a reanalysis of data to determine a life-time limit for ^{53}Cu. A comparison with data for ^{52}Ni and ^{54}Zn combined with EPAX calculations allows to determine a life-time limit of this nucleus of 130 ns which, in a simple barrier penetration model, yields a one-proton separation energy of −350 keV. This experimental limit is in agreement with a prediction from the latest Atomic Mass Evaluation and with other model predictions.

[AME-12 lists S(p)(^{53}Cu) = -1860(1060) from systematics]

2013Bl04 state: "The isotope ^{54}Cu was found unbound in a GSI experiment where ^{58}Ni fragmentation was used to produce proton-rich radioisotopes [4]. However, nothing could be said about ^{53}Cu".

Ref. 4: 1994Bl10, formal publication of 1994BlZW (or 1993Bl.A) report

^{53}Cu, ^{195}Os: short datasets but much effort

- ^{53}Cu: this nuclide has now been revised/updated in ENSDF by B. Singh and A. Chakraborty: June 11, 2013.

- ^{195}Os: revision for ENSDF is about done. There is quite a bit of current experimental data. It will soon be submitted for inclusion in ENSDF, and possible nuclide publication (~3 pages) in NDS by B. Singh, M. Birch, J. Flegenheimer, Z. Schaedig, and M. Thoennessen.

Moral of the stories

- For ENSDF evaluation, avoid copying blindly from other compiled or so-called evaluated databases. Double-checking of literature and follow-up papers is important.
- Thorough search of bibliography, NSR may not be the only source. For old (prior to 1965 or so) references, NSR is not that useful since key-words are mostly missing. For older references on radioactive isotopes, TOI-1978 is useful.
- Communicate with original authors. Over the years I have found most researchers are willing to help you out, some will even go out of their way to first find and then re-analyze several years old data.