

Article

First Direct Mass Measurement of ^{184}Ta and Surrounding Nuclei

Jinn Ming Yap ^{1,2} , Peter Schury ^{2,*} , Sarah Naimi ³ , Gemma Bartram ⁴ , Alison M. Bruce ⁵ , Navodhayan Chandrakumar ^{4,6} , Jiulong Chen ⁷ , James G. Cubiss ⁸ , Siddharth Doshi ⁵ , Sunil Dutt ⁹, Jilehu Gada ⁹ , Song Guo ⁹ , Yoshikazu Hirayama ² , George Hudson-Chang ^{4,6} , Sota Kimura ² , Filip G. Kondev ¹⁰ , Gregory Lane ¹¹ , Jenny Lee ¹ , Guangshun Li ⁹ , Yury Litvinov ^{12,13} , Zhong Liu ^{9,14} , Pengcong Ma ⁹ , Sorin Pascu ^{4,15} , Zsolt Podolyák ⁴ , Aiko Takamine ¹⁶ , Michiharu Wada ⁹ , Philip Walker ⁴ , Hiroshi Watanabe ⁷  and Yutaka X. Watanabe ² 

- ¹ Department of Physics, The University of Hong Kong, Pokfulam, Hong Kong, China; jmyap@connect.hku.hk (J.M.Y.); jleehc@hku.hk (J.L.)
- ² Wako Nuclear Science Center, Institute of Particle and Nuclear Studies, High Energy Accelerator Research Organization (KEK), Wako City 351-0198, Saitama, Japan; yoshikazu.hirayama@kek.jp (Y.H.); kimuraso@post.kek.jp (S.K.); yutaka.watanabe@kek.jp (Y.X.W.)
- ³ IJCLab, CNRS/IN2P3, Université Paris-Saclay, 91405 Orsay, France; sarah.naimi@ijclab.in2p3.fr
- ⁴ Department of Physics, University of Surrey, Guildford GU2 7XH, UK; g.bartram@surrey.ac.uk (G.B.); george.hudson-chang@riken.jp (G.H.-C.); s.pascu@surrey.ac.uk (S.P.); z.podolyak@surrey.ac.uk (Z.P.); p.walker@surrey.ac.uk (P.W.)
- ⁵ School of Architecture, Technology and Engineering, University of Brighton, Brighton BN2 4AT, UK; s.doshi1@uni.brighton.ac.uk (S.D.)
- ⁶ RIKEN Nishina Center, 2-1 Hirosawa, Wako City 351-0198, Saitama, Japan
- ⁷ School of Physics, Beihang University, Beijing 100191, China; jlchen@buaa.edu.cn (J.C.); hiroshi@ribf.riken.jp (H.W.)
- ⁸ School of Physics, Engineering and Technology, University of York, York YO10 5DD, UK; james.cubiss@york.ac.uk
- ⁹ Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China; sunilduttamu@impcas.ac.cn (S.D.); gadajilehu22@mails.ucas.ac.cn (J.G.); gs@impcas.ac.cn (S.G.); ligs@impcas.ac.cn (G.L.); liuzhong@impcas.ac.cn (Z.L.); mapengcong22@mails.ucas.ac.cn (P.M.); michiharu.wada@kek.jp (M.W.)
- ¹⁰ Physics Division, Argonne National Laboratory, Lemont, IL 60439, USA; kondev@anl.gov
- ¹¹ Department of Nuclear Physics, RSPHys, Australian National University, Canberra 2601, Australia; gregory.lane@anu.edu.au
- ¹² GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstraße 1, D-64291 Darmstadt, Germany; y.litvinov@gsi.de
- ¹³ Institut für Kernphysik, Universität zu Köln, Zùlpicher Str. 77, D-50937 Köln, Germany
- ¹⁴ School of Nuclear Science and Technology, University of Chinese Academy of Sciences, Beijing 100049, China
- ¹⁵ Horia Hulubei National Institute for R&D in Physics and Nuclear Engineering, R-77125 Bucharest-Magurele, Romania
- ¹⁶ Department of Physics, Kyushu University, 744 Motooka, Nishi-ku, Fukuoka City 819-0395, Fukuoka, Japan; takamine.aiko.329@m.kyushu-u.ac.jp
- * Correspondence: schury@post.kek.jp

Abstract

Mass measurements of isotopes around the $A = 184$ region near stability were performed at the KEK Isotope Separation System within the RIKEN Nishina Center, Japan. The isotopes were produced via multi-nucleon transfer reactions with a ^{136}Xe beam of 7.2 MeV/u impinging on a natural tungsten target. We report the first direct mass measurement of ^{184}Ta , showing a 62(27) keV deviation compared to previous indirect measurements.

Keywords: MRTOF-MS; MNT; nuclear mass



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1. Introduction

The nuclear mass is a fundamental property of atomic nuclei, enabling the study of nuclear structure and nucleosynthesis in astrophysics. Systematic studies of nuclear masses can reveal nuclear deformations and shell evolution from separation energy calculations [1,2]. These masses are also crucial inputs for nucleosynthesis calculations, elucidating the origins and abundance of naturally occurring isotopes [3].

Nuclear mass measurements are broadly categorized into direct and indirect methods. Indirect methods determine masses based on the reaction or decay Q-value, requiring prior knowledge of the nuclear transformation, including energy levels and decay modes. In comparison, direct methods are generally considered to be more accurate as mass determinations based on measurements of the orbital or cyclotron frequency, or the time-of-flight, do not require *a priori* knowledge about the nuclear reaction and decay schemes. For example, mass measurements of ^{184}Ir from $Q_{\beta+}$ measurements [4,5] were shown to have a large discrepancy (>600 keV) against newer storage ring mass measurements [6,7], of which the latter is accepted in the Atomic Mass Evaluation [8].

Direct mass spectroscopy methods such as Penning traps, storage rings and TOF- $\beta\rho$ spectrometry have tradeoffs between precision and measurement duration [9,10]. As mass measurements extend into more exotic regions, short life-times and low production yields have motivated the development of the multi-reflection time-of-flight mass spectrographs (MRTOF-MS) [11,12].

In this work we present the mass measurements of five isotopes, $^{178,180}\text{Re}$, $^{183,184}\text{Ir}$, and ^{184}Ta , complementing the investigation of K-isomers in this region [13,14]. The production of isotopes of interest in this work was done using multi-nucleon transfer (MNT) reactions which are characterized by the exchange of many nucleons between the target and projectile nuclei and have been gaining popularity in radioactive beam facilities such as GANIL [15], JAEA [16], GSI [17], ANL [18], and NEXT [19]. The KEK Isotope Separation System (KISS) was commissioned at the RIKEN Nishina Center [20–22] to produce a high-purity radioactive isotope beam of otherwise difficult-to-produce nuclides via MNT reactions for nuclear studies. The MNT products are captured and neutralized by a gas cell, and then selectively re-ionized in a two-photon resonant laser method. To perform high-precision mass measurements, an MRTOF-MS has been installed [23–25] downstream of a mass-selective dipole magnet. Mass measurements were performed using the KISS MRTOF-MS, alongside an evaluation of these new measurements' effect on the AME2020 mass surface.

2. Materials and Methods

Figure 1 shows the experimental setup at KISS. A primary beam of ^{136}Xe with an energy of 7.2 MeV/u and an intensity of 50 pA was accelerated by the RIKEN Ring Cyclotron (RRC). The primary beam impinges on a rotating natural tungsten target (28% ^{186}W) with a thickness of 9.627 mg/cm². The isotopes of interest were produced as target-like fragments (TLFs) in the MNT reactions and exited the target at a large grazing angle.

An 88 kPa argon-filled “doughnut-shaped” gas cell is positioned downstream from the target. The gas cell is designed such that the unreacted primary beam is allowed to pass through a 15 mm diameter central aperture without entering the gas cell. The target-side of the gas cell is constructed with a 5 μm thick Polyimide window. This allows the MNT products, which are emitted from the target at a large angle, to be captured by the gas cell while minimizing ionization of the argon gas. The MNT products are neutralized in the gas cell and subsequently transported to a two-step, two-color resonant laser ionization zone to selectively ionize nuclei with proton number, Z , of interest [26].

The atoms of the isotope of interest were first excited using an excimer-pumped dye laser (XeCl , $\lambda = 308$ nm, Lambda Physik, LPX240i). The laser wavelength, λ_1 , was tuned

such that it corresponded to the energy of the excited state of the element of interest. The six elements of primary interest—Pt, Ir, Os, Re, W, and Ta—were each excited at their known λ_1 values. The excited atoms were then exposed to laser radiation from a second excimer laser of the same make and model, but with a fixed wavelength of $\lambda_2 = 308$ nm, which ionizes them to the singly charged state.

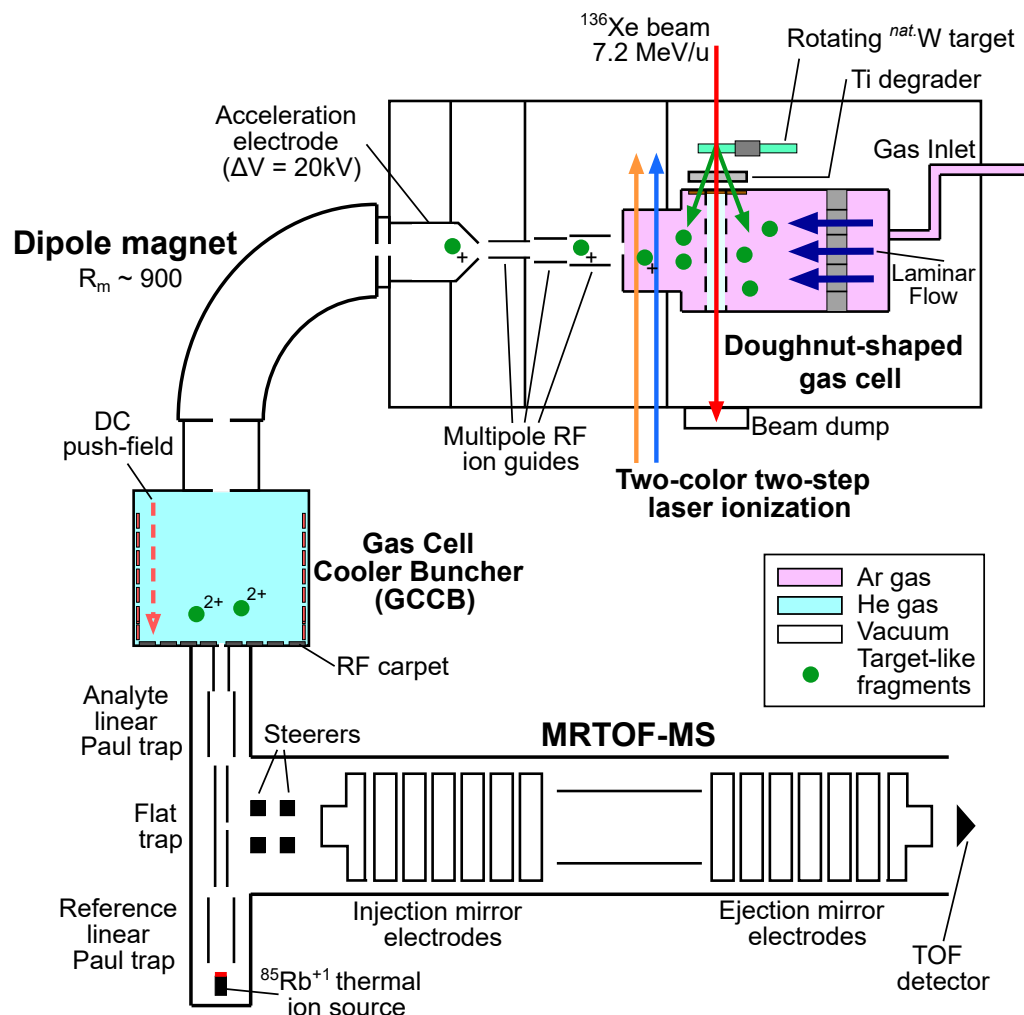


Figure 1. Schematic of the KISS setup modified from [24]. A ^{136}Xe beam impinges on a rotating tungsten target. Target-like fragments from the resulting MNT reactions are captured in the argon-filled doughnut-shaped gas cell (pink) and transported to the helium-filled GCCB (cyan) before mass measurement at the MRTOF-MS.

The ions were extracted through a small exit nozzle and transported to high-vacuum by a set of multipole radio-frequency (RF) ion guides through two differential vacuum pumping sections; the gas cell and differentially pumped chamber were on a 20 kV high-voltage platform. Upon reaching the end of the differentially pumped chamber, the ions were accelerated to 20 keV and sent through a dipole magnet mass separator section with a mass-resolving power of $m/\Delta m \sim 900$. The mass-separated ions are then guided into a 120 Pa helium-filled, windowless Gas Cell Cooler Buncher (GCCB) to be re-thermalized [23]. Vacuum pressure upstream of the windowless GCCB is maintained by a differential pumping system [27]. Initial energetic collisions with helium atoms in the GCCB strip additional electrons from the ions; subsequent low-energy interactions with the helium atoms result in doubly charged ions [28] being dominant. These ions are guided by a DC field onto an RF carpet that guides them to a small extraction hole. The ions are transported sequentially

via a quadrupole RF ion guide (QPIG) and a linear Paul trap to a flat trap [29], which cools the ions with <0.1 Pa He gas.

The TOF was measured starting with the injection of ions into the MRTOF-MS from the flat trap. The ions were then reflected between electrostatic mirrors of the MRTOF-MS for about 600 laps, and ejected onto an ion detector to complete a single measurement. The TOF of $^{85}\text{Rb}^+$ ions from a thermal alkali ion source, installed on the opposite end of the flat trap, was measured concomitantly with the ions of interest (analyte ions) [30]. The absolute mass values of the isotopes of interest ($Ae/q \approx 184/2$) was determined by the single-reference method [31],

$$\frac{m_a - (q_a/e)m_e}{q_a} = \frac{m_r - (q_r/e)m_e}{q_r} \rho^2, \quad \rho = \frac{t_a - t_0}{t_r - t_0}, \quad (1)$$

where m, q , and t are the atomic mass, charge, and TOF of the analyte and reference ions denoted by the subscripts a and r respectively. The flat trap ejection delay, t_0 , was empirically determined from the measurement of ρ from $^{85}\text{Rb}^+$ and $^{184}\text{W}^{2+}$ ions.

3. Results and Discussion

The TOF spectra centered on mass numbers $A = 178, 180, 183$, and 184 were analyzed. The TOF of observed ions was extracted after peak fitting using the hyper-EMG function [32] with identical shape parameters for all isobars. We were able to identify ions of eight nuclides, of which mass values of five isotopes ($^{178,180}\text{Re}^{2+}$, $^{183,184}\text{Ir}^{2+}$, $^{184}\text{Ta}^{2+}$) were determined as shown in Table 1. The remaining three ($^{178}\text{Hf}^{2+}$, $^{183,184}\text{W}^{2+}$) were used as isobaric references, except in the case of $^{180}\text{Re}^{2+}$ as it was the only $A = 180$ nuclide which could be clearly identified; instead it was measured with the reference ion $^{85}\text{Rb}^+$ and t_0 determined using $^{184}\text{W}^{2+}$.

All measured masses are in agreement with AME2020 values within $1\text{-}\sigma$ except for ^{184}Ta , which was previously determined only indirectly via the β^- decay endpoint energy [33]. Resonant laser ionization was used as a confirmation of the $^{184}\text{Ta}^{2+}$ peak as shown in Figure 2. By comparing the TOF spectra of $A = 184$ isobars with resonant ionization targets of tungsten and tantalum, a large increase in $^{184}\text{Ta}^{2+}$ ions was observed in the latter spectrum. On the other hand, the remaining peaks show no significant difference between resonant ionization targets as they originate from non-resonant excitations, which have lower cross-sections. Thus, we report here the first direct measurement of ^{184}Ta with a mass deviation from AME2020 of $\Delta m = 62(27) \text{ keV}/c^2$. We also report improved precision of ^{184}Ir compared to both direct and indirect measurements from the literature [4,5,34]. A comparison of mass excesses from this work against AME2020 [8,35] is shown in Figure 3.

Table 1. Masses of five isotopes were experimentally determined in this work. Ion species are listed with the reference ion used, the square of the time-of-flight ratio subtracted by one, ($\rho^2 - 1$), experimental mass excess (ME_{exp}), mass excess from AME2020 (ME_{AME}), and the difference ($\Delta\text{ME} = \text{ME}_{\text{exp}} - \text{ME}_{\text{AME}}$).

Analyte	Reference	$(\rho^2 - 1) \times 10^6$	$\text{ME}_{\text{exp}} \text{ (keV}/c^2)$	$\text{ME}_{\text{AME}} \text{ (keV}/c^2)$	$\Delta\text{ME} \text{ (keV}/c^2)$
$^{178}\text{Re}^{2+}$	$^{178}\text{Hf}^{2+}$	41.0224 (0.1805)	−45,635.8 (29.9)	−45,653.5 (27.9)	17.7 (40.9)
$^{180}\text{Re}^{2+}$	$^{85}\text{Rb}^+$	59,634.0930 (0.0444)	−45,839.6 (70.2)	−45,837.4 (21.4)	−2.2 (73.4)
$^{183}\text{Ir}^{2+}$	$^{183}\text{W}^{2+}$	35.9953 (0.2207)	−40,231.5 (37.6)	−40,202.1 (24.7)	−29.4 (45.0)
$^{184}\text{Ta}^{2+}$	$^{184}\text{W}^{2+}$	17.0888 (0.0419)	−42,777.5 (8.0)	−42,839.5 (26.0)	62.0 (27.2)
$^{184}\text{Ir}^{2+}$	$^{184}\text{W}^{2+}$	35.4087 (0.0716)	−39,638.2 (12.3)	−39,610.9 (27.9)	−27.3 (30.5)

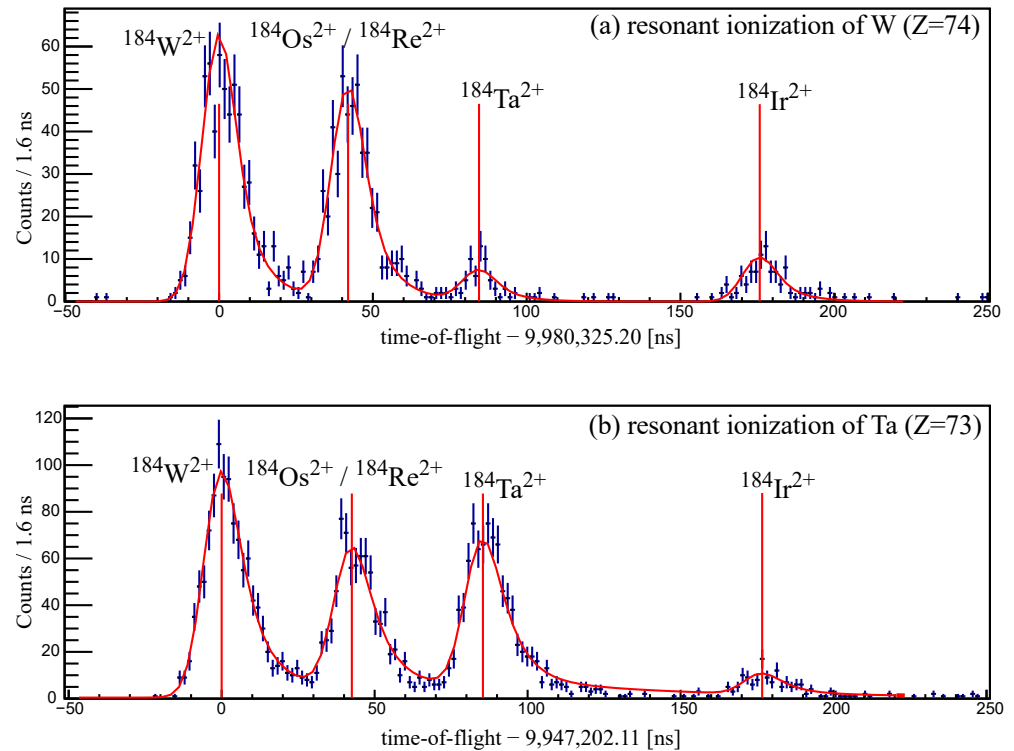


Figure 2. TOF spectra of $A = 184$ isobars with (a) laser resonant ionization of tungsten at 601 laps and (b) laser resonant ionization of tantalum at 599 laps. Four peaks are fitted as shown in red. A large increase in $^{184}\text{Ta}^{2+}$ ions is observed in (b), confirming the tantalum peak. ^{184}Re and ^{184}Os cannot be distinguished due to a small mass excess difference of $\sim 30 \text{ keV}/c^2$.

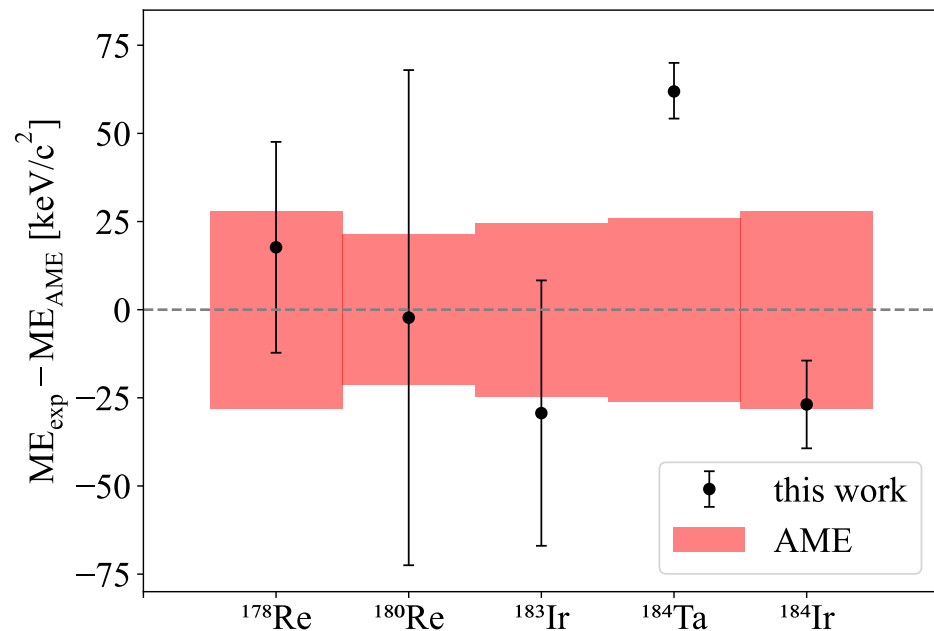


Figure 3. Mass excess differences between this work and those listed in AME2020 [35]. The sections shaded in red indicate the AME2020 mass uncertainties.

After extracting the mass values from our experimental data, we incorporated them into the Atomic Mass Evaluation (AME) network. Many of the masses we measured had already been determined using other methods. However, due to the high precision of our data, they had a significant impact on the network. In the AME network, nuclides are categorized as primary data if they are checked by other data; otherwise, they are considered secondary data.

In Table 2, we illustrate how our data influenced the primary dataset. With the exception of ^{184}Ta , which is classified as secondary data, all our measurements contribute to the AME primary data. Notably, in AME2020, only ^{183}Ir was considered a primary nucleus. However, after integrating our data, additional primary nuclei – $^{178,180}\text{Re}$ and ^{184}Ir – were introduced into the network.

Table 2. Influences of the isotopes measured in this work on the AME dataset, with the largest two influences (Infl.) and equations (Eq.) in the mass evaluation. Mass excesses (MEs) shown here are obtained from AME calculations after incorporating experimental values from this work.

Iso.	ME (keV/c ²)	Infl.	Eq.	Infl.	Eq.
^{178}Re	−45,645 (20)	53.3	$^{178}\text{Re-u}$	46.7	$^{178}\text{Re-}^{178}\text{Hf}$
^{180}Re	−45,838 (20)	91.4	$^{180}\text{Re-u}$	8.6	$^{180}\text{Re-}^{85}\text{Rb}$
^{183}Ir	−40,211 (21)	54.2	$^{183}\text{Ir-u}$	30.5	$^{183}\text{Ir-}^{183}\text{W}$
^{184}Ta	−42,777 (8)				
^{184}Ir	−39,634 (11)	84.1	$^{184}\text{Ir-}^{184}\text{W}$	15.9	$^{184}\text{Ir-u}$

Most of our measured masses align with previous measurements, except for ^{184}Ta . The mass of ^{184}Ta was previously determined through the β^- decay chain Hf-Ta-W, with a precision of 26 keV/c² [33]. However, since our new measurement has a higher precision of 8 keV/c², it now serves as the dominant data for determining the mass of ^{184}Ta . Additionally, our data influences the mass of ^{184}Hf , as it remains linked to ^{184}Ta through β^- decay. This adjustment resulted in a shift of approximately 60 keV/c² for ^{184}Hf , from −41,500(40) keV/c² to −41,440(30) keV/c², with slightly improved precision.

Two-neutron separation energy values, S_{2n} , were calculated to determine the impact of the updated mass values on the mass surface with

$$S_{2n}(Z, N) = m(Z, N - 2) - m(Z, N) + 2m_n \quad (2)$$

where m_n is the neutron mass and $m(Z, N)$ is the mass of a nuclide with Z protons and N neutrons. Figure 4 shows the S_{2n} of Hf, Ta and Ir isotope chains compared against several theoretical mass models, namely the microscopic mass model, HFB32 [36], as well as macroscopic–microscopic models FRDM12 [37] and WS4-RBF [38]. The S_{2n} along the Ta isotope chains were reproduced by the WS4-RBF model, though it should be noted that the updated S_{2n} of ^{184}Ta ($N = 111$) moved away from the predicted values of all mass models. The Ir isotope chain is well described by both WS4-RBF and FRDM12 as opposed to the HFB32 model; however, updated values of $^{183,184}\text{Ir}$ ($N = 106, 107$) suggest a trend towards FRDM12. The remaining Hf isotope chain is inconclusive as the trend is largely reproduced by the WS4-RBF model with the exception of ^{186}Hf ($N = 114$). This inconsistency may be due to the lack of direct mass measurements such as ^{184}Hf which was measured via the β^- decay endpoint energy [39]. Thus, more work is needed to evaluate the masses in this region with higher precision.

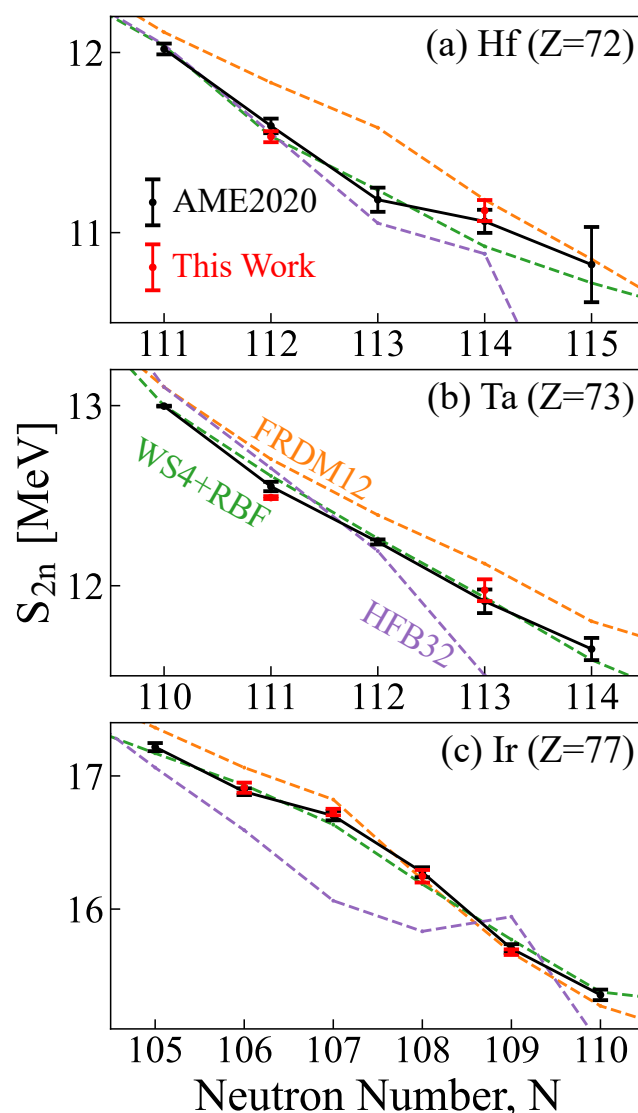


Figure 4. Two-neutron separation energy values of the (a) Hf, (b) Ta and (c) Ir isotope chains. The black line indicates values taken from AME2020, while the red markers indicate S_{2n} values with newly evaluated masses of ^{184}Hf , ^{184}Ta , and $^{183,184}\text{Ir}$. Colored dashed lines represent theoretical mass models listed in panel (b).

4. Conclusions

In conclusion, the masses of several isotopes spanning $A = 178$ through $A = 184$ were measured with high precision by MRTOF-MS as summarized in Table 1. The mass of ^{184}Ta is directly measured for the first time in this work, shifting the evaluated mass by approximately $60 \text{ keV}/c^2$ against AME2020. This shift is also reflected in the mass of ^{184}Hf . Additionally, the resulting shift in S_{2n} value of ^{186}Hf away from WS4+RBF towards FRDM12 highlights the importance of further precise mass measurements with direct methods for isotopes in this region.

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Data Availability Statement: Dataset available on request from the authors.

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