

Alpha-decay half-life of Hafnium isotopes reinvestigated by a semi-empirical approach*

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Abstract - New estimates of partial α -decay half-life, $T_{1/2}$, for $^{156-162,174,176}\text{Hf}$ isotopes by a semi-empirical, one-parameter model are given. The used model is based on the quantum mechanical tunneling mechanism through a potential barrier, where the Coulomb, centrifugal and overlapping components to the barrier have been considered within the spherical nucleus approximation. This approach enables to reproduce, within a factor 2, the measured $T_{1/2}$ of ground-state to ground-state (gs-gs) α -transitions for the artificially produced $^{156-162}\text{Hf}$ isotopes. Half-life predictions for α -transitions from the ground-state of $^{159,161}\text{Hf}$ isotopes to the first gamma-excited level of $^{155,157}\text{Yb}$ isotopes are reported for the first time. The model also provides $T_{1/2}$ -values of $(2.43 \pm 0.28) \times 10^{16}$ a and $(1.47 \pm 0.19) \times 10^{20}$ a for the naturally occurring ^{174}Hf and ^{176}Hf isotopes, respectively, in quite good agreement with a number of estimates by other authors. In addition, the present methodology indicates that $^{174,176}\text{Hf}$ isotopes exhibit α -transition to the first gamma-excited level of their daughter Ytterbium isotopes which half-lives are found $(0.9 \pm 0.1) \times 10^{18}$ a and $(0.72 \pm 0.08) \times 10^{22}$ a, respectively, with a chance of being measured by improved α -detection and α -spectrometry methods available nowadays.

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1 Introduction

Early studies by Henry Moseley and Georges Urbain showed that there were four missing elements between Aluminium and Gold in the 1914 Periodic Table. One of them was Element 72, discovered nine years later in *Hafnia* (the Latin name of Copenhagen) by George von Hevesy and Dirk Coster, which they called Hafnium (Authier, 2013).

Five naturally occurring Hafnium isotopes, *viz.*, $^{176-180}\text{Hf}$, were firstly identified by Aston in 1934 (Aston, 1934) followed by the discovery of ^{174}Hf by Dempster five years later (Dempster, 1939). Presently, the isotopic abundance of Hafnium reported in current Tables of Elements and Isotopes (Audi, 2017; Magill et al., 2018; Holden et al., 2019) is 0.16, 5.26, 18.60, 27.28, 13.62, and 35.08 atoms percent for $^{174,176-180}\text{Hf}$ isotopes, respectively.

The first radioactive Hafnium isotope, ^{181}Hf —a β^- -active nuclide of half-life 42.39 d— was identified in 1935 by Hevesy and Levi, who bombarded a Hafnium sample with neutrons forming the reaction $^{180}\text{Hf}(n,\gamma)$, and measured the β^- -rays after chemical separation (Hevesy and Levi, 1935). From 1949 on, thirty-one new radioactive Hafnium isotopes have been identified as product nuclides in a number of nuclear reactions (for detailed information see (Gross and Thoennessen, 2012)). Six of these radioisotopes are partially α -active ($^{157-162}\text{Hf}$), competing with the electron capture process, and only one (^{156}Hf) is totally α -active. Their α -half-lives are found in the range 24 ms–5.7 d (Audi, 2017).

It happened that, in 1959, Riezler and Kauw observed for the first time an alpha activity exhibit by the naturally occurring ^{174}Hf isotope, and they measured its half-life as 4.3×10^{15} a (Riezler and Kauw, 1959). Two years later, the measured value $(2.0 \pm 0.4) \times 10^{15}$ a was obtained by Macfarlane and Kohman (Macfarlane and Kohman, 1961), and this result has been until now the one quoted in all nuclear data compilations (*see note added in proof*).

In their recent review article on long-lived alpha emitters Belli *et al.* (Belli et al., 2019) have reported for ^{174}Hf isotope the measured α -decay half-life as $T_{1/2}^\alpha = (2.0 \pm 0.4) \times 10^{15}$ a, and theoretical values of 3.5×10^{16} a and 0.74×10^{17} a. For the other naturally occurring $^{176-180}\text{Hf}$ isotopes $T_{1/2}^\alpha$ is quoted in the range $\sim 10^{20}$ – 10^{46} a, *i.e.*, very difficult to be measured (or even impossible), and such Hf isotopes are considered stable. A point of special attention discussed in the paper mentioned above is the case for $^{178m2}\text{Hf}$ isomer ($T_{1/2} = 31$ a), for which case the question of its partial α -decay half-life is not yet completely elucidated, deserving careful investigation (see (Belli et al., 2019) and references therein).

Quite recently, Ukrainian and Belgian researchers conducted a series of experiments to detect alpha decays of $^{174,176-180}\text{Hf}$ isotopes to the first excited state of $^{170,172-176}\text{Yb}$ isotopes, and of ^{179}Hf to the ground state of ^{175}Yb (Danevich et al., 2020). The measurements were carried out at a depth of 225 m at the HADES underground laboratory (Belgium) by using high-purity Germanium detectors of ultra-low background. Since no decays were recorded after two and a half months of counting, only lower limits for the half-life in the range $\sim 10^{15}-10^{18}$ a could be reported (Danevich et al., 2020).

Apart from numerous experiments of measuring of α -decay half-life of naturally occurring and artificially produced isotopes along the Nuclide Chart, in the last forty years or so a large number of investigations have been carried out towards a deeper understanding of the phenomenon of alpha decay using different phenomenological and microscopic models, most of them allowing to calculate the half-lives (Poenaru et al., 1980; Poenaru et al., 1986; Buck et al., 1991; Poenaru and Greiner, 1991; Buck et al., 1993; García et al., 2000; Royer, 2000; Moustabchir and Royer, 2001; Duarte et al., 2002; Delion and Sandulescu, 2002; Xu and Ren, 2004; Dupré and Bürvenich, 2006; Medeiros et al., 2006; Li et al., 2010; Denisov and Khudenko, 2009; Santhosh et al., 2011; Poenaru et al., 2011; Poenaru et al., 2013; Bao et al., 2014; Qian and Ren, 2014; Denisov et al., 2015; Wang et al., 2015). Additionally, over the decades several empirical or semi-empirical formulas have been proposed (Poenaru et al., 1980; Poenaru et al., 1986; Buck et al., 1991; Buck et al., 1993; García et al., 2000; Royer, 2000; Moustabchir and Royer, 2001; Dupré and Bürvenich, 2006; Medeiros et al., 2006; Li et al., 2010; Denisov and Khudenko, 2009; Poenaru et al., 2011; Qian and Ren, 2014; Denisov et al., 2015; Wang et al., 2015; Poenaru and Ivascu, 1983; Alex Brown, 1992; Parkhamenko and Sobiczewski, 2005; Poenaru et al., 2007; Ni et al., 2008; Royer, 2010; Akrawy and Poenaru, 2017), and eventually they have been useful to promptly estimate the half-lives, what have motivated researches to new experiments to detect yet unobserved α -decay cases. Quite recently, an improved and simple empirical α -decay half-life formula by Deng et al. has given more reliable predictions for a large number of both favored and unfavored α -transition cases, as well as for α -decay chains of superheavy elements (Deng, Zhang and Royer, 2020).

The understanding of the decay features of Hafnium isotopes is not only an issue of scientific interest. In effect, one must also consider that the Element Hafnium plays a role in several fundamental technological fields, from nuclear and reactor physics to metallurgy. Moreover, some naturally occurring long-living Hafnium isotopes are being used in space science to investigate the history of terrestrial planets and confirm timescales determined by the Re/Os, Pt/Os, Sm/Nd and U/Pb systems (Kinny and Maas, 2003; Kleine et al., 2009; Iizuka et al., 2015).

One of the most important clocks used to constrain timescale of planetary differentiation is

the ^{176}Lu - ^{176}Hf radioactive system (Scherer et al., 2001). Recently, the chemical analysis of the $^{176}\text{Hf}/^{177}\text{Hf}$ ratio in a rare meteorite allowed to dating the Earth's first crust formation at about 4.6×10^9 years ago (Bast et al., 2017). However, despite the importance of ^{176}Hf isotope, up to now no data have been reported regarding its decay and ^{176}Hf is currently considered a stable isotope (Holden et al., 2019). It should be mentioned that the discrepancies found in the initial Hafnium-isotope ratios, originating mainly from uncertainties, or even lack, of decay data make now difficult a precise dating of early stages of solar system by Hf-based chronometry.

Based on the considerations above, and since there is no publication specifically dedicated to the half-life of the alpha-emitting Hf isotopes, we thought it worthwhile to undertake a reinvestigation on alpha activities of both artificially produced and naturally occurring Hafnium isotopes.

In the present investigation we have applied a semi-empirical, one-parameter model to obtain the half-lives of all α -active Hafnium isotopes, as well as to predict decay rates for new, yet unobserved α -transitions of these isotopes. This approach has been introduced by us a few years ago (Tavares et al., 2005) to explain the rare α -activity found in monoisotopic, natural bismuth (^{209}Bi) that was announced by de Marcillac *et al.* (de Marcillac et al., 2003). Then the model was successfully applied to Platinum isotopes (Tavares et al., 2006; Tavares and Medeiros, 2011), ^{147}Sm isotope (Tavares and Terranova, 2018), Osmium isotopes (Tavares and Terranova, 2020), in a systematic study of all α -emitter nuclides (Medeiros et al., 2006), and also extended to cases of radioactivity by emission of one- and two-proton from nuclides close to the proton dripline (Medeiros et al., 2007; Tavares and Medeiros, 2018), and to cluster radioactivity as well (Tavares and Medeiros, 2012). We believe the new α -decay data which will arise from the methodology described in (Tavares and Medeiros, 2012) can motivate experimental groups toward novel, updated measured α -decay half-life data for Hafnium isotopes.

2 A simple calculation model for heavy-particle radioactivity

In the last fifteen years or so, we have developed and applied a routine calculation model to estimate half-life of different cases and modes of nuclear decay by heavy-particle emission such as one-proton and simultaneous two-proton emission modes, α -decay, and cluster radioactivity (the emission of nuclear particles heavier than the α -particle, like ^{14}C , ^{20}O , ^{23}F , and others, also known as exotic radioactivity). The basis for the present method is the current quantum mechanical tunneling mech-

anism through a Coulomb-plus-centrifugal-plus-overlapping potential barrier constructed within the spherical nucleus approximation.

Since the fundamentals of the model have been previously reported to some details (Tavares et al., 2005; Tavares and Medeiros, 2012; Tavares and Terranova, 2018), here we present such a methodology in a summary form only. In what follows subscripts P, D, and C refer to parent emitter nucleus, daughter product nucleus, and emitted nuclear heavy-particle (or nuclear cluster), respectively. In addition, in the formulas that follow, lengths and distances are expressed in fm ($1 \text{ fm} = 10^{-13} \text{ cm}$), masses in u ($1 \text{ u} = 1.660539040 \times 10^{-24} \text{ g}$), energies in MeV ($1 \text{ MeV} = 1.602176565 \times 10^{-6} \text{ erg}$), and time in year ($1 \text{ a} = 3.1557 \times 10^7 \text{ s}$). Accordingly, the half-life has been computed as

$$\log_{10} T_{1/2} = \tau = \tau_{\text{fa}} + \tau_{\text{ov}} + \tau_{\text{se}}, \quad (1)$$

where

$$\tau_{\text{fa}} = -29.5 + \log_{10} \left[a \cdot (\mu_0/Q_a)^{1/2} \right] \quad (2)$$

is the term related to the frequency of assaults of the α -particle on the potential barrier,

$$\tau_{\text{ov}} = g \cdot \tau_1, \quad \tau_1 = 0.19 \cdot (c - a) \cdot (\mu_0 \cdot Q_a)^{1/2} \cdot H(x, y) \quad (3)$$

is the contribution due to the overlapping barrier region, g is the adjustable parameter of the model, and

$$\tau_{\text{se}} = \tau_2^{\text{coul}} + \tau_2^{\text{cent}} = 0.27358 Z_C \cdot Z_D \cdot (\mu_0/Q_a)^{1/2} \cdot F(x, y) \quad (4)$$

is the one corresponding to the external, separation barrier region. The Coulomb and centrifugal terms in (4) can be separated as

$$\tau_2^{\text{coul}} = 0.27358 Z_C \cdot Z_D \cdot (\mu_0/Q_a)^{1/2} \cdot F(0, y), \quad \text{and} \quad (5)$$

$$\tau_2^{\text{cent}} = \tau_2^{\text{coul}} \cdot [F(x, y)/F(0, y) - 1]. \quad (6)$$

Here, $a = R_P - R_C$ is the difference between the radius of the parent nucleus and the cluster radius; the length $2a$ represents the linear displacement of the cluster between two successive assaults to the barrier. The quantity $c = R_D + R_C$ defines the configuration at contact of the preformed fragments (daughter product nucleus and emitting cluster), so that the length $c - a = 2R_C - (R_P - R_D)$ gives the width of the overlapping barrier region.

$$\mu_0 = [m_C^{-1} + m_D^{-1}]^{-1} \quad (7)$$

is the reduced mass of the produced nuclides, where the m 's represent the nuclear (rather than atomic) masses, the values of which are calculated as

$$m_i = A_i - Z_i \cdot m_e + \left(\Delta M_i + k Z_i^\beta \right) / F, \quad i = \text{P, D, C}, \quad (8)$$

where $m_e = 0.54857990907 \times 10^{-3}$ u is the electron rest mass, $\Delta M'_i$'s are the atomic mass-excess values (expressed in MeV), kZ_i^β represents the total binding energy of the Z electrons in the atom, and $F = 931.4940038$ MeV/u is the mass-energy conversion factor.

In this way, the total available energy for the disintegration $P \rightarrow D + C$ system, $Q_a = m_P - (m_D + m_C)$, is obtained as

$$Q_a = \Delta M_P - (\Delta M_D + \Delta M_C) + S, \quad S = kZ_P^\beta - (kZ_D^\beta + kZ_C^\beta), \quad (9)$$

where S denotes the screening effect to the nuclei caused by the surrounding electrons. The k - and β -values come from an analysis of calculated data for electron binding energies of neutral atoms by Huang *et al.* (Huang et al., 1976). From the analysis it is obtained

$$k = 8.7 \times 10^{-6} \text{ MeV}, \quad \beta = 2.517, \quad \text{for nuclei of } Z \geq 60, \quad \text{and} \quad (10)$$

$$k = 13.6 \times 10^{-6} \text{ MeV}, \quad \beta = 2.408, \quad \text{for nuclei of } Z < 60. \quad (11)$$

Finally, the functions $H(x, y)$ and $F(x, y)$ contain the dependence upon mutual angular momentum, ℓ (if any), which comes from the nuclear spin (\mathbf{J}) and parity (π) conservation laws ($\mathbf{J}_P = \mathbf{J}_D + \mathbf{J}_C + \boldsymbol{\ell}$, $\pi_P = \pi_D \cdot \pi_C \cdot (-1)^\ell$) to the decay process. These functions are written as

$$H(x, y) = (x + 2y - 1)^{1/2}, \quad (12)$$

$$F(x, y) = x^{1/2}/(2y) \cdot \ln \left\{ [x^{1/2} \cdot H(x, y) + x + y] / (x + y^2)^{1/2} \right\} + \arccos \left\{ [1 - (y - 1)/(x + y^2)^{1/2}] / 2 \right\}^{1/2} - H(x, y)/(2y), \quad (13)$$

and the quantities x and y are defined as

$$x = 20.9008 \cdot \ell(\ell + 1)/(\mu_0 \cdot c^2 \cdot Q_a), \quad y = (1/2)Z_C Z_D e^2 / (c \cdot Q_a), \quad (14)$$

where $e^2 = 1.43996444$ MeV·fm is the square of the electronic elementary charge.

Expressions (12) and (13) are valid for all values of $x \geq 0$ and $y > 1/2$. In cases for which $\ell = 0$ it results $x = 0$, $\tau_2^{\text{cent}} = 0$, and hence

$$\tau_1 = 0.19 \cdot (c - a) \cdot [\mu_0 \cdot Q_a \cdot (u^{-1} - 1)]^{1/2}, \quad \text{and} \quad (15)$$

$$F(0, y) = \arccos u^{1/2} - [u \cdot (1 - u)]^{1/2}, \quad u^{-1} = 2y. \quad (16)$$

The single, semi-empirical g parameter of the model emerges from a combination of exponents p and q in the power functions that describe the reduced mass, $\mu(s)$, and the potential barrier, $V(s)$,

respectively, in the overlapping region ($a \leq s \leq c$; s is the distance of separation between the centers of the forming product nuclides), as

$$g = [1 + (p + q)/2]^{-1}, \quad 0 < g \leq 2/3. \quad (17)$$

The origin of this expression for parameter g has been explained in Ref. (Tavares et al., 2005).

The g -value depends on the source of input data for both nuclear mass and radius, on the adopted values for the different physical constants, and, of course, on the measured half-life data of a given set of alpha-decay cases. Once a mass table and/or a nuclear radius parametrization were chosen, subsequent half-life evaluations should be done using these sources for mass- and radius-values, as well as the physical constants from which the g -value was obtained. Parameter g is related to the strength of the spectroscopic factor, *i.e.*, the cluster preformation probability, whose value is given by $10^{-g \cdot \tau_1}$.

3 Application to α -decay of Hafnium isotopes

The parent, α -emitting Hafnium isotopes ($Z = 72$) transform to daughter product Ytterbium isotopes ($Z = 70$) and ${}^4\text{He}$ clusters ($Z = 2$), hence the quantity $C = 0.27358 \cdot Z_C \cdot Z_D = 38.3012$ which appears in expressions (4) and (5) is a constant for all α -decay cases. Also, it results that the Q_α -value and its uncertainty, δQ_α , for the cases under examination reduce to

$$Q_\alpha = \Delta M_P - \Delta M_D - 2.396817 \text{ MeV}, \quad \delta Q_\alpha = [(\delta \Delta M_P)^2 + (\delta \Delta M_D)^2]^{1/2}, \quad (18)$$

in which atomic mass-excess values are those quoted in the mass tables by Wang *et al.* (The AME2016 atomic mass evaluation (II) (Wang et al., 2017)); the screening contribution to Q_α -value amounts to $S \sim 28.1$ keV, and variables y and u take the values

$$y = 100.7975108/(c \cdot Q_\alpha) \quad \text{and} \quad u = c \cdot Q_\alpha/201.5950216. \quad (19)$$

We choose to evaluate the nuclear radius values for the parent, R_P , and daughter, R_D , nuclides by following the finite range droplet model of atomic nuclei as is described by Möller *et al.* (Möller et al., 1995), where the spherical approximation of the nuclear volume has been adopted (see also (Myers, 1977)). This nuclear radius parametrization has been updated recently by Möller *et al.* (Möller et al., 2016). The new radius data have been obtained from a significantly improved treatment that took into account more accurate experimental ground-state nuclear mass data. The expressions that

enable to calculate the average equivalent root-mean-square radius of the proton and neutron density distributions are now read as

$$R_i \equiv \langle Q_{Z,A} \rangle = (Z/A)Q_p + (1 - Z/A)Q_n, \quad i = P, D, \quad (20)$$

where the equivalent radii, Q_j ($j = p, n$), are obtained from

$$Q_j = R_j \left(1 + 5/(2R_j^2)\right), \quad (21)$$

in which R_j represents the sharp radii for the proton and neutron density distributions ($j = p, n$, respectively), the values of which are given by

$$R_p = r_0(1 + \langle \epsilon \rangle) \cdot [1 - (2/3) \cdot (1 - Z/A) \cdot (1 - 2Z/A - \langle \delta \rangle)] \cdot A^{1/3}, \quad (22)$$

$$R_n = r_0(1 + \langle \epsilon \rangle) \cdot [1 + (2/3) \cdot (Z/A) \cdot (1 - 2Z/A - \langle \delta \rangle)] \cdot A^{1/3}. \quad (23)$$

Here, $r_0 = 1.16$ fm, and the values for the quantities $\langle \delta \rangle$ and $\langle \epsilon \rangle$ are reported in (Möller et al., 2016) as

$$\langle \delta \rangle = (1 - 2Z/A + 0.0048626 \cdot Z/A^{2/3}) / (1 + 2.5304666/A^{1/3}), \quad (24)$$

$$\langle \epsilon \rangle = 0.854167 \cdot \exp(-0.988A^{1/3}) - 0.1896936/A^{1/3} + 0.2229167 \cdot \langle \delta \rangle^2 + 0.0031034 \cdot Z^2/A^{4/3}. \quad (25)$$

In the case for Hafnium and Ytterbium isotopes the ratio $r_i = R_i/A_i^{1/3}$ ($i = P, D$) obtained according to the parametrization reported above reveals a small, but important, decrease when one passes from ^{152}Yt ($r_D = 1.223$ fm) to ^{176}Hf ($r_P = 1.217$ fm). These results indicate a clear degree of nuclear compressibility, making, therefore, the simple expression $R = \langle r \rangle \cdot A^{1/3}$ not valid in estimating nuclear radii in the whole range of mass number.

Concerning the alpha-particle radius, it has been adopted the value $R_C \equiv R_\alpha = (1.62 \pm 0.01)$ fm, in accordance to the α -particle radius-value derived from the charge density distribution measured by Sick *et al.* (Sick et al., 1976) in electron scattering experiments on ^4He target. Excellent reproducibility of alpha-decay data was attained using the above mentioned R_α -value coupled with a simple Gamow's-like model applied to a large number (more than three hundred cases) of measured alpha-decay half-lives covering the mass-number interval $106 \leq A \leq 264$ (Medeiros et al., 2006). From the radius parametrization reported above the width of the overlapping barrier region, $c - a$, amounts to ~ 3.18 fm.

3.1 Determination of parameter g

The first task is to find the value of parameter g and its uncertainty, δg . Then we can proceed to calculate the half-lives of all α -transitions of Hafnium isotopes, compare results with the measured

values, and make predictions for new α -decay cases.

The input data for all α -emitting Hafnium isotopes to be used in finding the value of parameter g are shown in table 1. It is seen that in all cases $\ell = 0$, hence $\tau_{se} = \tau_2^{\text{coul}}$. Then, starting from eqs. (1) and (3), one has

$$g^e = (\tau^e - \tau_{fa} - \tau_2^{\text{coul}}) \cdot \tau_1^{-1}, \quad (26)$$

$$\delta g^e = [(\delta\tau^e)^2 + (\delta\tau_{fa})^2 + (\delta\tau_2^{\text{coul}})^2 + (g^e \cdot \delta\tau_1)^2]^{1/2} \cdot \tau_1^{-1}, \quad (27)$$

where superscript ‘e’ indicates that g^e -values come from experiment. In (26) and (27), τ_{fa} , τ_1 , and τ_2^{coul} are given by eqs. (2), (15), and (5), respectively, combined with eq. (16), and $\tau^e = \log T_{1/2}^\alpha$, where $T_{1/2}^\alpha$ is expressed in a. The uncertainties associated to τ^e , τ_{fa} , τ_1 , and τ_2^{coul} are evaluated as

$$\delta\tau^e = 0.43429 \cdot (\delta T_{1/2}^\alpha / T_{1/2}^\alpha), \quad \delta\tau_{fa} = 0.21715 \cdot (\delta Q_\alpha / Q_\alpha), \quad (28)$$

$$\delta\tau_1 = 0.018 \cdot (c - a)^2 \cdot (\mu_0 / \tau_1) \cdot \delta Q_\alpha, \quad (29)$$

$$\delta\tau_2^{\text{coul}} = \left\{ \tau_2^{\text{coul}} / 2 + 2.7 \cdot [\mu_0 \cdot c \cdot (1 - u)]^{1/2} \right\} \cdot (\delta Q_\alpha / Q_\alpha). \quad (30)$$

Results for nine values of g^e and their uncertainties are listed in table 1 (last column). It is seen that nuclear data for the artificially produced $^{156-162}\text{Hf}$ isotopes lead to positive g^e -values in the interval 0.047–0.150, thus compatible with the range of g -values allowed by the model. However, in the case for ^{174}Hf isotope both the two measured half-life values led to negative g^e -values. Since g^e must necessarily be a positive number, the present model predicts a preliminary lower limit of $\sim 0.8 \times 10^{16}$ a for the half-life of ^{174}Hf .

In the case of nuclides of the same isotopic sequence that exhibit alpha transitions with the same value of ℓ (in this case $\ell = 0$) we can assume a single value of g , $\langle g^e \rangle$, to obtain the calculated α -decay half-lives of Hafnium isotopes. The $\langle g^e \rangle$ -value has been evaluated by minimizing the quantity

$$D = \sum (\tau^c - \tau^e)^2, \quad \tau^c = \tau_{fa} + \langle g^e \rangle \cdot \tau_1 + \tau_2^{\text{coul}}, \quad (31)$$

that is, by solving for $\langle g^e \rangle$ from the condition $dD/d\langle g^e \rangle = 0$, which gives

$$\langle g^e \rangle = \left(\sum \tau^e \cdot \tau_1 - \sum \tau_{fa} \cdot \tau_1 - \sum \tau_2^{\text{coul}} \cdot \tau_1 \right) / \sum \tau_1^2. \quad (32)$$

The estimated uncertainty associated to $\langle g^e \rangle$ is calculated as $\delta\langle g^e \rangle = (1/N) \cdot [\sum (\delta g^e)^2]^{1/2}$. In this way, one obtains

$$\langle g^e \rangle = 0.0919 \pm 0.0077 (2\sigma). \quad (33)$$

The present value found for $\langle g^e \rangle$ -parameter compares well with the one found previously by us in an analysis of Platinum isotopes ($\bar{g} = 0.105$) (Tavares and Medeiros, 2011), and with the value 0.103

obtained quite recently in an investigation on Osmium isotopes as well (Tavares and Terranova, 2020). In addition, since the chance to find the α -particle at the nuclear surface of the product nucleus (α -preformation probability) is given by $p_\alpha = 10^{-g \cdot \tau_1}$, and considering that for all the examined Hafnium isotopes the average value of τ_1 has been found as $\tau_1 \approx 5.3 \pm 0.1$, it follows that $p_\alpha \approx 32\%$. This result is much the same as the one found in α -decay of ^{147}Sm isotope (Tavares and Terranova, 2018) and for a number of Osmium isotopes (Tavares and Terranova, 2020).

3.2 Calculated half-life values and comparison with experimental data

To obtain the calculated half-life values it suffices to insert back into eq. (1) the semi-empirical value $\langle g^e \rangle = 0.0919 \pm 0.0077$ found for the single parameter of the model, *i.e.*,

$$\tau^c = \tau_{\text{fa}} + \langle g^e \rangle \cdot \tau_1 + \tau_2^{\text{coul}}, \quad (34)$$

and its uncertainty given by

$$\delta\tau^c = [(\delta\tau_{\text{fa}})^2 + (\tau_1 \cdot \delta\langle g^e \rangle)^2 + (\langle g^e \rangle \cdot \delta\tau_1)^2 + (\delta\tau_2^{\text{coul}})^2]^{1/2}. \quad (35)$$

Then,

$$T_{1/2}^c = \exp(\tau^c \cdot \ln 10) \quad \text{and} \quad \delta T_{1/2}^c = T_{1/2}^c \cdot \delta\tau^c \cdot \ln 10. \quad (36)$$

The present formalism has been applied, besides to the gs–gs α -transitions of $^{156-162,174,176}\text{Hf}$ isotopes, also to investigate the possibility of transitions to the first gamma-excited level of daughter Ytterbium isotopes. Since for these last transitions $\ell \neq 0$, the contribution due to the centrifugal term

$$\tau_2^{\text{cent}} = \tau_2^{\text{coul}} \cdot [F(x, y)/F(0, y) - 1], \quad \delta\tau_2^{\text{cent}} = \delta\tau_2^{\text{coul}} \cdot [F(x, y)/F(0, y) - 1] \quad (37)$$

should be added to (34) and (35), respectively. The routine calculation proceeds as before, this time by introducing the appropriate Q_α -values,

$$Q_\alpha^* = Q_\alpha^{\text{gs}} - E_\gamma, \quad (38)$$

where E_γ is the gamma quantum energy of the level.

Results and comparison with experimental data are presented in table 2. It is seen that the naturally occurring $^{174,176}\text{Hf}$ isotopes, besides showing α -transitions to the ground state of $^{170,172}\text{Yb}$ isotopes, respectively, exhibit also α -transitions to the first gamma quantum excited level of the respective daughter Ytterbium isotopes. The present half-life predictions for these latter α -transitions

are compatible with the experimental lower limits obtained by Danevich *et al.* (Danevich et al., 2020), and much closer to the theoretical predictions of Denisov *et al.* (Denisov et al., 2015).

The present calculation model predicts that the artificially produced $^{159,161}\text{Hf}$ isotopes can also yield α -particles which populate the first excited level of their respective daughter $^{155,157}\text{Yb}$ isotopes with non-negligible intensities (last column of table 2).

In figure 1 we depict the α -decay schemes which emerge from the present half-life calculations for the α -emitting $^{159,161,174,176}\text{Hf}$ isotopes. For all cases examined, calculated partial or total α -decay half-lives are given in the seventh and tenth columns, respectively, of table 2, and a comparison with experimental values in the ninth column. These latter data show that the ratio of calculated to measured values does not exceed a factor 2, except for ^{174}Hf isotope (a factor ~ 13). Very probably such a discrepancy noted in the case for ^{174}Hf isotope could be caused by the significant difference of ~ 63 keV between the α -particle energy ($E_\alpha = (2.50 \pm 0.03)$ MeV) measured at the time of the experiment (Macfarlane and Kohman, 1961) and the currently accepted value of $E_\alpha = 2.437$ MeV (Wang et al., 2017). Calculations have indeed indicated that such a difference of ~ 63 keV leads to a difference of about one order of magnitude in half-lives. The fact is that a number of estimates made by other authors (73 % of values listed in table 3) points to a half-life for ^{174}Hf isotope in the range $(1.2\text{--}5.5)\times 10^{16}$ a, that is, about one order of magnitude greater than the measured values. As for ^{176}Hf isotope the half-life estimates listed in Table 3 point to a value in the range $\sim (0.55\text{--}5.0)\times 10^{20}$ a (*see note added in proof*).

3.3 Linearization of α -decay half-life data

All α -decay data presented in the precedent subsection can be displayed in a form that allows one to describe the half-life values essentially as a linear function of the total energy available for decay, the Q_α -value.

Starting from eqs. (1)–(4), we define the “reduced” calculated, τ_r^c , and “reduced” experimental, τ_r^e , half-lives by subtracting from the half-life τ (eq. (1)) all contributions other than the Coulomb one, *i.e.*,

$$\tau_r^c = \tau^c - (\tau_{\text{fa}} + \langle g^e \rangle \cdot \tau_1 + \tau_2^{\text{cent}}) \quad \text{and} \quad \tau_r^e = \tau^e - (\tau_{\text{fa}} + \langle g^e \rangle \cdot \tau_1 + \tau_2^{\text{cent}}), \quad (39)$$

in such a way that

$$\Delta\tau_r = \tau_r^c - \tau_r^e = \tau^c - \tau^e = \Delta\tau. \quad (40)$$

Here, the assumption is that $\langle g^e \rangle$, the single parameter of the model, takes the same value for all

α -decay cases, *i.e.*, $\langle g^e \rangle$ is a constant in all α -transitions from the ground state, α -emitting Hafnium isotopes.

Equation (39) means that $\tau_r^c = \tau_2^{\text{coul}} = C \cdot z$, where $C = 38.3012$ and

$$z = (\mu_0/Q_\alpha)^{1/2} \cdot \arccos u^{1/2} - [u \cdot (1 - u)]^{1/2}, \quad u = c \cdot Q_\alpha/201.5950216. \quad (41)$$

Thus, τ_r^c *versus* z gives a straight line of slope C , and, from (40), $\tau_r^e = C \cdot z - \Delta\tau$.

Figure 2 displays all “reduced” τ_r^c and τ_r^e half-life values obtained following the present methodology for the thirteen α -transition cases listed in Table 2. As one can see, the reduced experimental half-life data (full circles) appear very closely to the straight line, except for $^{174}\text{Hf}(\text{gs}) \rightarrow ^{170}\text{Yb}(\text{gs}) + \alpha$ transition, whose τ_r^e -value deviates by ~ 1 order of magnitude.

4 Final comments and conclusion

The semi-empirical, one-parameter calculation model outlined in the precedent sections enabled us to reproduce quite well the measured α -decay half-life values of the artificial $^{156-162}\text{Hf}$ isotopes, and, moreover, to make reliable half-life predictions for the naturally occurring $^{174,176}\text{Hf}$ isotopes.

For gs–gs α -transitions of $^{174,176}\text{Hf}$ isotopes the evaluated half-lives were found $(2.43 \pm 0.28) \times 10^{16}$ a and $(1.47 \pm 0.19) \times 10^{20}$ a, respectively. The same methodology has been also employed to predict cases of gamma-quanta accompanying α -decay of $^{174,176}\text{Hf}$ isotopes and possible of being accessed experimentally. For α -decay of $^{174,176}\text{Hf}$ isotopes to the first excited gamma-quantum level of their daughter Ytterbium isotopes the present model yielded half-life values of $(0.9 \pm 0.1) \times 10^{18}$ a and $(0.72 \pm 0.08) \times 10^{22}$ a, respectively.

As a whole, the evaluated data suggest a chance for the α -decay half-lives of the naturally occurring $^{174,176}\text{Hf}$ isotopes to be measured by means of the improved experimental methods of α -detection and α -spectrometry available nowadays (Beeman et al., 2012; Beeman et al., 2013; Münster et al., 2014) (*see note added in proof*).

Some possibility of being detected seems indeed to exist for the Ytterbium isotopes descending from $^{174,176}\text{Hf}$ parent isotopes. Taking for instance a ~ 900 -g $\text{Hf}(\text{WO}_4)_2$ scintillating bolometer crystal, during six months of activity the number of alpha particles from $^{174}\text{Hf} \rightarrow ^{170}\text{Yb}^{\text{gs}}$ ($E_\alpha = 2.44$ MeV) and $^{174}\text{Hf} \rightarrow ^{170}\text{Yb}^{\text{1es}}$ ($E_\alpha = 2.35$ MeV) decays could be $\sim 1.7 \times 10^4$ and ~ 490 , respectively, whereas from $^{176}\text{Hf} \rightarrow ^{172}\text{Yb}^{\text{gs}}$ ($E_\alpha = 2.20$ MeV) and $^{176}\text{Hf} \rightarrow ^{172}\text{Yb}^{\text{1es}}$ ($E_\alpha = 2.13$ MeV) decays, the emission of ~ 100 and ~ 2 α 's would be expected.

In addition, the adopted methodology enabled to point out, for the first time, cases of α -transitions of the artificial $^{159,161}\text{Hf}$ isotopes to the first γ -excited level of $^{155,157}\text{Yb}$ isotopes. The data obtained by the present evaluation allowed us to propose decay schemes for the α -decay processes of $^{159,161,174,176}\text{Hf}$ isotopes that populate the first excited level of their respective Ytterbium daughters (Fig. 1).

An insight into the present model can help in rationalizing the general features shown by the α -decay process of Hafnium isotopes. The decay constant, λ , is expressed as $\lambda = \lambda_0 \cdot S \cdot P$, where

$$\lambda_0 = a^{-1} \cdot [Q_\alpha / (2\mu_0)]^{1/2} = 2.192 \times 10^{29} \cdot a^{-1} \cdot (Q_\alpha / \mu_0)^{1/2} \text{ a}^{-1} \quad (42)$$

represents the number of assaults on the potential barrier per unit of time, $S = 10^{-\tau_{\text{ov}}} = 10^{-(g^e)\tau_1}$ is the α -particle preformation probability, and $P = 10^{-\tau_{\text{se}}}$ is the penetrability factor through the external barrier region created by the separated fragments. Since for all α -decay cases examined in the present study the quantity λ_0 (in units of 10^{29} a^{-1}) varies in the narrow interval ~ 0.3 – 0.5 , and τ_{ov} in the ~ 0.48 – 0.52 one, it follows that λ (or $T_{1/2} = (\ln 2)/\lambda$) is essentially dictated by P , the penetrability factor through the Coulomb-plus-centrifugal separation region. In view of the fact that τ_{se} -values vary in a very large range (~ 19 – 50) going from ^{156}Hf to ^{176}Hf , the decay constant is expected to vary by 31 orders of magnitude, and the variation of the α -decay half-life for the α -emitting Hafnium isotopes (Fig. 2) finds a rational explanation.

In concluding, we want to stress the importance to dispose of reliable experimental data on α -decaying Hafnium isotopes in order to confirm the predictions of the present model and, in the case of ^{174}Hf isotope, to elucidate the discrepancies found with old measurements.

On the one hand, as regards the yet undetected α -activity of ^{176}Hf isotope, descendent from ^{176}Lu through β^- -decay process with a half-life of (37.13 ± 0.16) Ga (Söderlund et al., 2004), the half-life value obtained by the present calculation could help in resolving still controversial chronology of events that occurred in the earliest solar system and defining astrophysical sites of complex-element nucleosynthesis as well. Experiments aiming at determining the half-life of ^{176}Hf isotope are strongly suggested.

Additionally, the radioactive behavior of Hafnium isotopes may have significant implications also for other research fields. As a matter of fact, the use of element Hafnium is presently not limited only to studies of nuclear physics and geo- and cosmo-chronology, or for applications in nuclear plants and in advanced metallurgy. This (tetravalent transition) metal is positioned at the forefront of several novel technological fields, *in primis* in nano-electronics, where Hafnium-based materials have emerged in the last years as designated dielectrics for future generation of devices (Choi et

al., 2011). As an example, HfO_2 is one of the most reliable among the high- k oxides proposed for replacing the conventional SiO_2 -gate in Si-based devices. But the integration of Hf-based components in nano-electronics could present fundamental limits, due to intrinsic α -radioactive decay of natural Hafnium.

For all these reasons it is clear that efforts in investigating α -decay of Hafnium isotopes must be increased. The task to measure half-lives for such long-term decaying isotopes is a challenging one, but it can be reasonably accomplished taking advantage of the new measurement techniques developed by experimental groups of excellence. We hope that the findings of the current research could provide a convincing starting point and trigger studies along this direction.

Note added in proof - An alternative experimental result for the α -decay half-life of ^{174}Hf isotope has been quite recently obtained by Caracciolo *et al.* (2020) as $(7.0 \pm 1.2) \times 10^{16}$ a, in good agreement with a number of calculated values (see Table 3).

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Table 1 - Nuclear data for α -emitting Hafnium isotopes ($Z = 72$) to determine parameter g of the present heavy-particle decay calculation model*.

Decay case		$T_{1/2}^\alpha$ (gs-gs) ^{a,b}	Q_α -value (MeV) ^c	Nuclear radii (fm) ^d		Reduced mass	
Parent (P)	Daughter (D)			R_P	R_D	μ_0 (u) ^e	g^e -value
$^{156}\text{Hf}(0^+) \rightarrow$	$^{152}\text{Yb}(0^+)$	(24 ± 1) ms	6.05666 ± 0.00381	6.580	6.528	3.89881	0.1205 ± 0.0046
$^{157}\text{Hf}(7/2^-) \rightarrow$	$^{153}\text{Yb}(7/2^-)$	(122 ± 1) ms	5.90818 ± 0.00306	6.598	6.540	3.89946	0.1499 ± 0.0024
$^{158}\text{Hf}(0^+) \rightarrow$	$^{154}\text{Yb}(0^+)$	(6.43 ± 0.16) s ^f	5.43287 ± 0.00272	6.604	6.552	3.90011	0.0971 ± 0.0031
$^{159}\text{Hf}(7/2^-) \rightarrow$	$^{155}\text{Yb}(7/2^-)$	(0.54 ± 0.01) min ^g	5.25319 ± 0.00267	6.617	6.565	3.90074	0.0757 ± 0.0029
$^{160}\text{Hf}(0^+) \rightarrow$	$^{156}\text{Yb}(0^+)$	(32.4 ± 0.5) min	4.92996 ± 0.00259	6.629	6.577	3.90137	0.1089 ± 0.0028
$^{161}\text{Hf}(7/2^-) \rightarrow$	$^{157}\text{Yb}(7/2^-)$	(3.93 ± 0.08) h	4.71040 ± 0.02408	6.641	6.590	3.90199	0.0494 ± 0.0253
$^{162}\text{Hf}(0^+) \rightarrow$	$^{158}\text{Yb}(0^+)$	(5.70 ± 0.13) d	4.44441 ± 0.00486	6.653	6.602	3.90261	0.0473 ± 0.0058
$^{174}\text{Hf}(0^+) \rightarrow$	$^{170}\text{Yb}(0^+)$	$(2.0 \pm 0.4) \times 10^{15}$ a	2.52262 ± 0.00226	6.799	6.750	3.90943	-0.1062 ± 0.0167
$^{174}\text{Hf}(0^+) \rightarrow$	$^{170}\text{Yb}(0^+)$	4.3×10^{15} a ^h					-0.0464 ± 0.0094

*Since $J_P^\pi = J_D^\pi$ in all cases, it follows that $\ell = 0$.

^a These are half-life values for ground-state to ground-state α -transitions.

^b Unless otherwise specified, values of $T_{1/2}^\alpha$ have been taken from (Audi, 2017).

^c Screening effect due to surrounding electrons taken into account (eq. (9)).

^d These are obtained following the Finite Range Droplet Model by Möller *et al.* (Möller et al., 2016).

^e This is given by $\mu_0 = (m_D^{-1} + m_\alpha^{-1})^{-1}$, where the m 's represent nuclear (rather than atomic) mass-values.

^f Quoted in (Reich, 2009).

^g Quoted in (Reich, 2005).

^h First measured value in 1959 (Riezler and Kauw, 1959).

Table 2 - Calculated partial and total α -decay half-lives of parent Hafnium isotopes for α -transitions to ground- and γ -excited-states of daughter Ytterbium isotopes.

Hafnium isotopes		Level of Yb-isotope ^a			α -transition		Partial half-life values (a) ^b			Total α -Half-life (calculated)	alpha intensity (%)
Parent	J_{P}^{π}	J_{D}^{π}	E_{γ} (keV)	Q_{α} (MeV)	ℓ	(this work)	measured	ratio ^c	(calculated)	(%)	
¹⁵⁶ Hf	0 ⁺	0 ⁺	0	6.05666	0	$(5.3 \pm 0.5)10^{-10}$	$(7.5 \pm 0.3)10^{-10}$	~ 1.4		100	
¹⁵⁷ Hf	7/2 ⁻	7/2 ⁻	0	5.90818	0	$(1.9 \pm 0.2)10^{-9}$	$(3.88 \pm 0.03)10^{-9}$	~ 2		100	
¹⁵⁸ Hf	0 ⁺	0 ⁺	0	5.43287	0	$(1.9 \pm 0.2)10^{-7}$	$(2.03 \pm 0.05)10^{-7}$	~ 1.1		100	
¹⁵⁹ Hf	7/2 ⁻	7/2 ⁻	0	5.25319	0	$(1.25 \pm 0.12)10^{-6}$	$(1.03 \pm 0.02)10^{-6}$	~ 1.2	$(1.16 \pm 0.11)10^{-6}$	92.8	
		9/2 ⁻	169.30	5.08389	2	$(1.6 \pm 0.5)10^{-5}$	————	————		7.2	
¹⁶⁰ Hf	0 ⁺	0 ⁺	0	4.92996	0	$(5.0 \pm 0.5)10^{-5}$	$(6.16 \pm 0.09)10^{-5}$	~ 1.2		100	
¹⁶¹ Hf	7/2 ⁻	7/2 ⁻	0	4.71040	0	$(7.6 \pm 2.4)10^{-4}$	$(4.5 \pm 0.1)10^{-4}$	~ 1.7	$(7.36 \pm 2.25)10^{-4}$	96.8	
		9/2 ⁻	205.5	4.50490	2	$(2.3 \pm 0.8)10^{-2}$	————	————		3.2	
¹⁶² Hf	0 ⁺	0 ⁺	0	4.44441	0	$(2.7 \pm 0.3)10^{-2}$	$(1.56 \pm 0.04)10^{-2}$	~ 1.7		100	
¹⁷⁴ Hf	0 ⁺	0 ⁺	0	2.52262	0	$(2.5 \pm 0.3)10^{16}$	$(2.0 \pm 0.4)10^{15}$	~ 13	$(2.43 \pm 0.28)10^{16}$	97.2	
		2 ⁺	84.25468	2.43836	2	$(0.9 \pm 0.1)10^{18}$	————	————		2.7	
¹⁷⁶ Hf	0 ⁺	0 ⁺	0	2.28230	0	$(1.5 \pm 0.2)10^{20}$	————	————	$(1.47 \pm 0.19)10^{20}$	98.0	
		2 ⁺	78.7427	2.20356	2	$(0.72 \pm 0.08)10^{22}$	————	————		2.0	

^a Data for J^{π} and E_{γ} have been taken from the most recent Nuclear Data Sheets publications of the corresponding A of the daughter Ytterbium isotope.

^b Other calculated and measured half-life data for gs-gs α -transitions of naturally occurring ^{174,176}Hf isotopes are reported in Table 3.

^c This is given by $10^{|\tau^c - \tau^e|}$.

Table 3 - Alpha-decay half-lives of the naturally occurring ^{174}Hf and ^{176}Hf isotopes ($Z = 72$).

Author(s)	Year	Half-life values (a)		Reference
		^{174}Hf (0.16 %)	^{176}Hf (5.26 %)	
Calculated				
Poenaru <i>et al.</i>	1986	0.63×10^{17}	5.0×10^{20}	Poenaru et al., 1986
Buck <i>et al.</i>	1991	3.8×10^{15}	$1.6 \times 10^{20\text{a}}$	Buck et al., 1991
Buck <i>et al.</i>	1993	2.3×10^{15}	—	Buck et al., 1993
García <i>et al.</i>	2000	3.7×10^{16}	$1.1 \times 10^{20\text{a}}$	García et al., 2000
Royer	2000	$1.3 \times 10^{16\text{a}}$	$0.63 \times 10^{20\text{a}}$	Royer, 2000
Duarte <i>et al.</i>	2002	5.5×10^{16}	$5.0 \times 10^{20\text{a}}$	Duarte et al., 2002
Xu and Ren	2004	$3.3 \times 10^{16\text{a}}$	3.0×10^{20}	Xu and Ren, 2004
Dupré and Bürvenich	2006	1.2×10^{16}	0.55×10^{20}	Dupré and Bürvenich, 2006
Medeiros <i>et al.</i>	2006	3.8×10^{16}	2.2×10^{20}	Medeiros et al., 2006
Denisov and Khudenko	2009	3.0×10^{16}	2.1×10^{20}	Denisov and Khudenko, 2009
Royer	2010	1.6×10^{16}	0.77×10^{20}	Royer, 2010
Qian and Ren	2014	4.43×10^{15}	2.25×10^{20}	Qian and Ren, 2014
Denisov <i>et al.</i>	2015	1.8×10^{16}	0.89×10^{20}	Denisov et al., 2015
Akrawy and Poenaru	2017	$2.0 \times 10^{16\text{b}}$	$1.0 \times 10^{20\text{b}}$	Akrawy and Poenaru, 2017
Tavares, Medeiros and Terranova	2020	2.5×10^{16}	1.5×10^{20}	this work
Experimental				
Porschen and Riezler	1956	$> 3 \times 10^{17}$	—	Porschen and Riezler, 1956
Riezler and Kauw	1959	4.3×10^{15}	—	Riezler and Kauw, 1959
Macfarlane and Kohman	1961	$(2.0 \pm 0.4) \times 10^{15}$	—	Macfarlane and Kohman, 1961

^a Extrapolated value.

^b Formula (3) with coefficients of set C in Ref. (Akrawy and Poenaru, 2017).

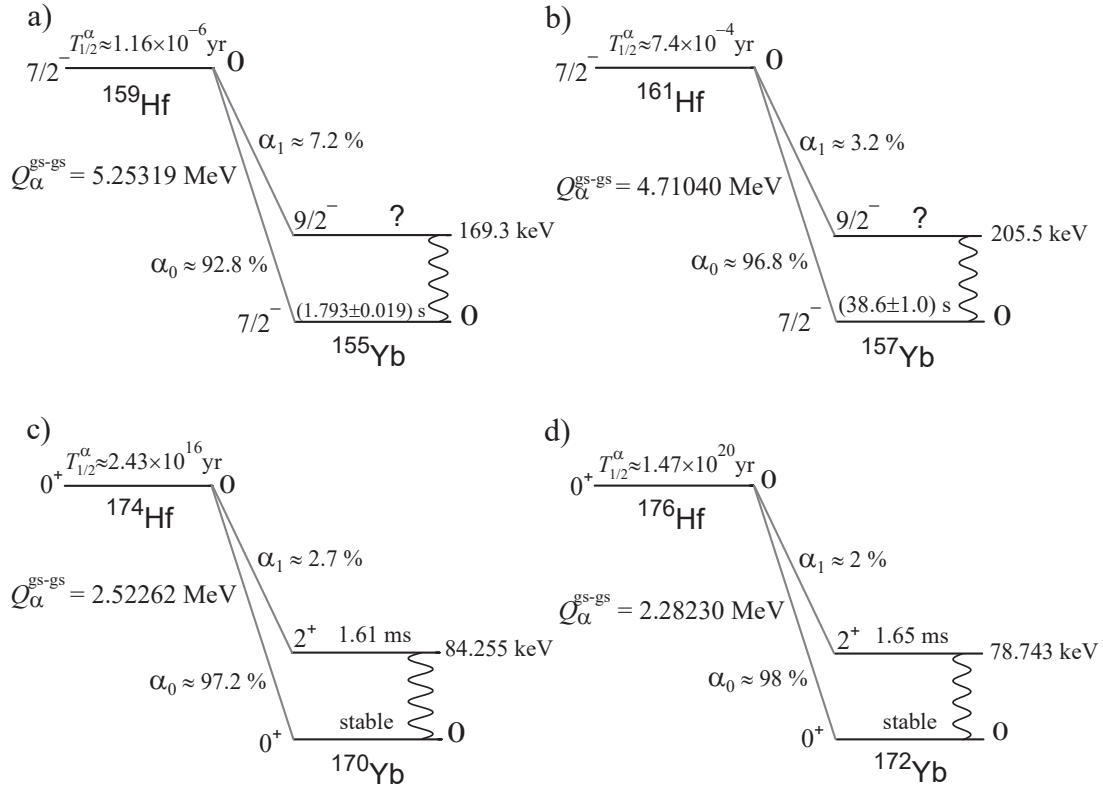


Fig. 1 Proposed α -decay schemes for ^{159}Hf (a), ^{161}Hf (b), ^{174}Hf (c), and ^{176}Hf (d) isotopes. Values reported for half-life and intensity of parent Hafnium isotopes have been obtained from the present α -decay methodology. Nuclear data of the levels and gamma quantum emissions of daughter Ytterbium isotopes are taken from (Reich, 2005), (Nica, 2016), (Baglin et al., 2018), and (Singh, 1995), respectively.

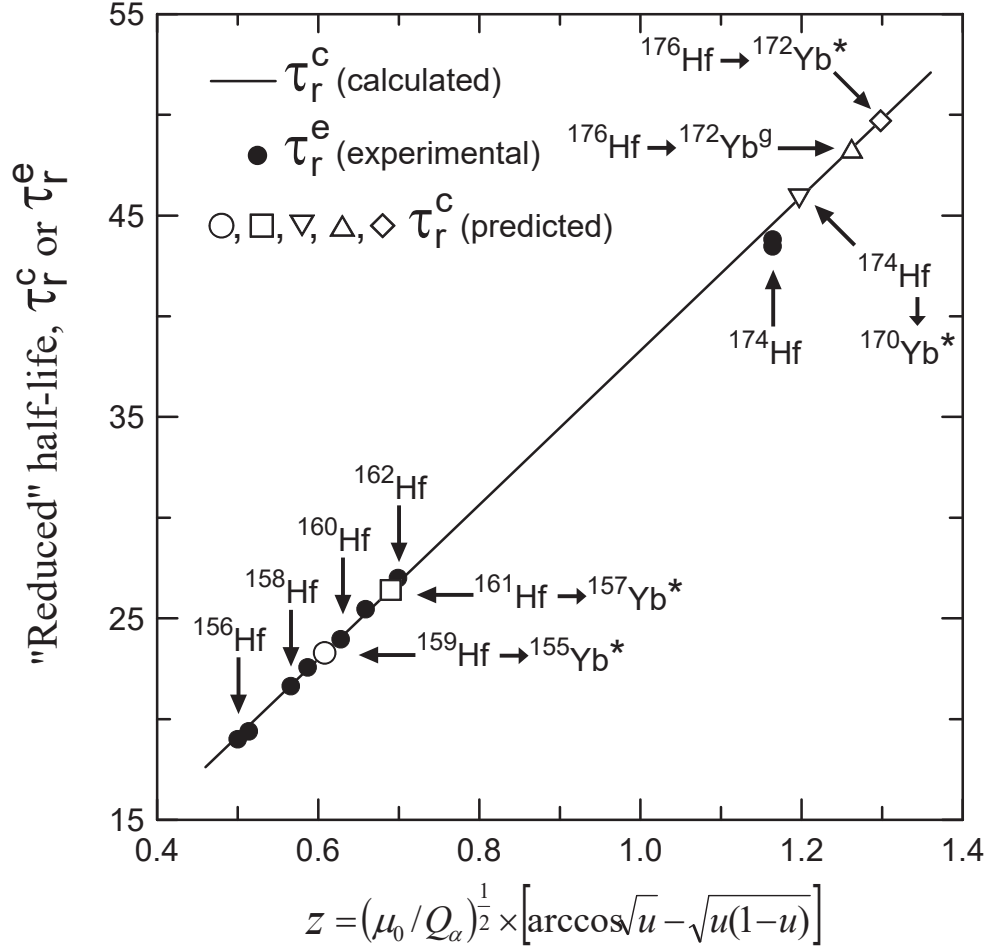


Fig. 2 Reduced half-life, $\tau_r = \tau_2^{\text{coul}}$ (eq. (39)), plotted against z for all cases of α -transitions of Hafnium isotopes. Calculated values are represented by the straight line $\tau_r^c = 38.3012 \cdot z$. Full circles represent experimental τ_r^e -values for all eight observed cases of gs-gs α -transitions; open symbols indicate yet unobserved gs-gs α -decay of ^{176}Hf isotope (Δ) and four cases (as indicated) of α -transitions to excited levels of daughter Yb isotopes which could emit γ quanta accompanying the α emission (the corresponding, predicted half-life-values are listed in Table 2).