

New evaluation of alpha decay half-life of ^{190}Pt isotope for the Pt-Os dating system

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Abstract - A semiempirical model based on the quantum mechanical tunnelling mechanism of alpha emission from nuclei has been used to evaluate the half-life of the Pt isotopes. For the important naturally occurring ^{190}Pt isotope, the radiogenic parent in the $^{190}\text{Pt}\rightarrow^{186}\text{Os}$ dating system, the model yielded a half-life value of $(3.7 \pm 0.3) \times 10^{11}$ y. This is comparable to $(3.2 \pm 0.1) \times 10^{11}$ y which was obtained in the last direct counting experiment to measure the alpha activity of ^{190}Pt (Tavares and Terranova, *Rad. Measur.* **27** (1997) 19). A literature survey of available alpha decay half-life values for ^{190}Pt isotope is also reported. The significant discrepancies found between data obtained by direct counting, indirect geological methods and different calculation models are analysed and discussed.

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The alpha decay of the ^{190}Pt isotope to the daughter ^{186}Os plays a fundamental role in the geochronometer based on the $^{190}\text{Pt}\rightarrow^{186}\text{Os}$ isotope system which has been used in many geochronological investigations as well as in geochemistry, cosmochemistry and space science [1–5]. Nowadays, as remarked by Walker *et al.* [2], the $^{190}\text{Pt}\rightarrow^{186}\text{Os}$ system may prove a valuable complementary method to the $^{187}\text{Re}\rightarrow^{187}\text{Os}$ system for dating some types of iron meteorites. However, radioisotopic age determination of both ores and meteorites are strongly dependent upon the various radionuclide half-life-values and the associated accuracies. As recognized by Begemann and coworkers [6] there is, in general, a strong need for accurate and unambiguous half-life evaluation for the isotopes to geochronological use.

In the case of the Pt-Os dating system, after the pioneering studies [2], further efforts towards effective applications have been hampered by difficulties arising from both the extremely small abundance ($\sim 0.013\%$ of atoms) and long half-life ($\gtrsim 10^{11}$ y) found for ^{190}Pt isotope. The resulting very low alpha activity ($\sim 1\text{--}2$ disintegration per minute and per gram of natural platinum) makes it very difficult indeed to obtain a precise and accurate experimental half-life value for ^{190}Pt by direct counting method. Other procedures

which have been employed in order to evaluate the ^{190}Pt half-life are geological comparison methods using other radionuclides (mainly ^{238}U) and semiempirical calculation models. Table 1 reports a review of the available ^{190}Pt half-life-data (both experimental and calculated values) obtained by using the different methods mentioned above. As one can see, the data listed in Table 1 span over a wide range of half-life values, and discrepancies like the ones shown in Table 1 can not be accounted for. Non-negligible differences are also found between the averaged values for each set of techniques. The value listed in the last line of the group of the direct counting methods has been obtained by a long-term exposure experiment (up to 2 y) carried out in the Gran-Sasso Underground Laboratory (INFN-LNGS, Italy) in order to redetermine the alpha decay half-life of the ^{190}Pt isotope. Details of the whole experiment have been reported in [15]. Basically, high-purity, metallic sheets of natural platinum were kept in close contact with plates of CR-39 nuclear-track detectors. The etched plates were analysed at CBPF/MCT (Rio de Janeiro, Brazil), and from the density of alpha-particle events $(4.91 \pm 0.15) \times 10^3 \text{ cm}^{-2}$, corresponding to a total amount of ^{190}Pt exposure of $6.2 \text{ mg}\cdot\text{h}\cdot\text{cm}^{-2}$, the value $(3.2 \pm 0.1) \times 10^{11} \text{ y}$ has been obtained for the half-life of ^{190}Pt isotope. However, this result is about one half of those reported by Macfarlane and Khoman [11] $((6.9 \pm 0.5) \times 10^{11} \text{ y})$ and Al-Bataina and Jänecke [14] $((6.65 \pm 0.28) \times 10^{11} \text{ y})$, although good agreement is found with the weighted average of $(3.5 \pm 0.3) \times 10^{11} \text{ y}$ resulting from calculation models (last line in Table 1).

Very recently, a research group detected for the first time alpha particles from the radioactive decay of natural bismuth (100% of ^{209}Bi isotope) by means of the scintillating bolometer technique with massive BGO ($\text{Bi}_4\text{Ge}_3\text{O}_{12}$) crystals [26]. The measured half-life was found as $(1.9 \pm 0.2) \times 10^{19} \text{ y}$, corresponding to an alpha activity equivalent to about ~ 12 disintegrations/h·kg, which represents the lowest natural alpha activity ever observed. The precise and accurate half-life value obtained in such spectacular experiment motivated us to develop a one-parameter model for alpha decay, based on the quantum mechanical tunnelling mechanism of penetration through a potential barrier, where the centrifugal and overlapping effects are taken into account [27]. This treatment enabled us to calculate and systematize the alpha decay half-lives of ground-state to ground-state transitions of mutual angular momentum $\ell = 5$ for all the possible alpha-emitter bismuth isotopes which can be produced by nuclear reactions. The detailed description of the model, and the input nuclear data for the routine calculation and half-life systematics, are reported in [27]. It was verified that the measured partial alpha decay half-life-values (covering 25 orders of magnitude) were reproduced by the proposed method quite satisfactorily (in some cases within 15%). In particular, the alpha decay half-life for the naturally occurring ^{209}Bi isotope was evaluated as $(1.0 \pm 0.3) \times 10^{19} \text{ y}$ [27], in substantial agreement with the experimental result $((1.9 \pm 0.2) \times 10^{19} \text{ y}$ [26]).

The proposed model has shown to be successfully applicable also to other isotopic sequences of alpha-emitter nuclides. In brief, the decay constant, λ , is calculated as

$$\lambda = \lambda_0 S_\alpha P_{se}, \quad S_\alpha = e^{-G_{ov}}, \quad P_{se} = e^{-G_{se}}, \quad (1)$$

where λ_0 is the frequency factor which represents the number of assaults on the barrier per unit of time, S_α is the alpha particle preformation probability at the nuclear surface (also known as the spectroscopic factor), G_{ov} is Gamow's factor calculated in the overlapping barrier region where the alpha particle drives away from the parent nucleus until the configuration at contact is reached (the quantity $S_\alpha = e^{-G_{ov}}$ can, therefore, be thought as the "arrival" of the alpha particle at the nuclear surface), P_{se} is the penetrability factor

through the external barrier region (*i.e.*, the Coulomb plus centrifugal potential barriers), and G_{se} is Gamow's factor calculated in this external, separation region which extends from the contact configuration up to the separation point where the total potential energy equals to the Q_α -value for decay. The quantity λ_0 is evaluated usually as $\lambda_0 = \frac{v}{2a}$, where v is the relative velocity of the fragments, and $a = R_p - R_\alpha$ is the difference between the radius of the parent nucleus and the alpha particle radius. Accordingly, if one expresses lengths in fm, masses in u, energies in MeV, and half-lives in y, the alpha decay half-life is obtained as

$$T_{1/2\alpha} = 3.17 \times 10^{-30} a \left(\frac{\mu_0}{Q_\alpha} \right)^{1/2} S_\alpha^{-1} \cdot P_{\text{se}}^{-1}, \quad (2)$$

where μ_0 represents the reduced mass of the disintegration system.

In cases for which the alpha transition goes from the ground-state of the parent nucleus to the ground-state of the daughter nucleus, and the transition has mutual angular momentum $\ell = 0$, the expressions for G_{ov} and G_{se} reduce to (the general expressions when $\ell \neq 0$ are given in [27]):

$$G_{\text{ov}} = 0.4374702(c - a)g (\mu_0 Q_\alpha)^{1/2} \left(\frac{1}{z} - 1 \right)^{1/2}, \quad (3)$$

$$G_{\text{se}} = 1.25988372 Z_d \left(\frac{\mu_0}{Q_\alpha} \right)^{1/2} \{ \arccos(z^{1/2}) - [z(1 - z)]^{1/2} \}, \quad (4)$$

in which

$$z = \frac{cQ_\alpha}{2Z_d e^2}, \quad e^2 = 1.4399652 \text{ MeV} \cdot \text{fm}. \quad (5)$$

Here, $c = R_d + R_\alpha$ is the separation of fragments in the contact configuration (R_d represents the radius of the daughter nucleus), and Z_d is the atomic number of the daughter nucleus. The quantity $c - a$ represents, therefore, the extension of the overlapping region. Finally, g is the adjustable parameter of the model, the value of which being thus determined from experimental data of partial alpha decay half-life of the alpha emitter nuclides of a given isotopic family. The evaluation of the quantities μ_0 , Q_α , R_α , R_p , and R_d has been described in detail in [27]. In this way, the best g -value is obtained by minimizing the quantity

$$\sigma = \left\{ \frac{1}{n - 2} \sum_{i=1}^n \left[\log_{10} \left(\frac{T_{1/2_i}^e}{T_{1/2_i}^c} \right) \right]^2 \right\}^{1/2}, \quad (6)$$

where $T_{1/2}^e$ and $T_{1/2}^c$ are the respective measured and calculated partial alpha decay half-life values, and n is the number of alpha disintegration cases in a given isotopic sequence. The g -value so obtained is then put back into the routine calculation of the model to calculate the $T_{1/2\alpha}$ -values for the n alpha emitter nuclides.

Results for new isotopic sequences other than those reported in [27] can be appreciated in Fig. 1. Despite the large range covered by both the half-life (~ 30 orders of magnitude) and atomic number ($Z = 52$ – 102) quantities, agreement between calculated and measured values can be considered rather satisfactory (in a number of cases the difference between each other is indeed very small ($\lesssim 10\%$)).

In the case of platinum ($Z = 78$) there are nineteen isotopes (including ^{190}Pt) which have experimental indication of total half-life and alpha-decay branching ratio for ground-state to ground-state transitions of mutual angular momentum $\ell = 0$, therefore their partial alpha decay half-lives are known [7,28] (see Table 2). By using the measured, partial alpha decay half-life-data for all the nineteen isotopes (Tables 1 and 2) as input to our semiempirical, one-parameter model, we found for the adjustable parameter of the model the value $g = 0.131$, corresponding to a minimum standard deviation (Eq. (6)) of $\sigma_{\min} = 0.201$. This best g -value is then used back into the routine calculation of the model to obtain the partial alpha decay half-life-values for all the nineteen alpha-active platinum isotopes. Again the experimental and calculated $T_{1/2\alpha}$ -values fit to each other quite satisfactorily (Fig. 2).

Of special interest to nuclear decay data compilation and geochronological use is the case for the ^{190}Pt isotope, for which the calculated alpha decay half-life value from the referred semiempirical model results to be $(3.7 \pm 0.3) \times 10^{11}$ y. This value (shown as open symbol in the inset of Fig. 2) is listed in the last line of Table 1 to allow an intercomparison between half-life data obtained by using different methods. Inspection of Table 1 shows a number of remarkable facts. First, the present calculated half-life-value agrees quite completely with the weighted average value of the direct counting experiments group, although it is about one half of the most accurate result by Al-Bataina and Jänecke [14]. Substantial agreement is also noted with the result $(3.2 \pm 0.1) \times 10^{11}$ y found in our previous measurement [15]. As pointed out above, this last value was obtained by direct counting of alpha-tracks produced during an experiment involving the greatest amount of exposure of ^{190}Pt (equivalent to $6.2 \text{ mg}\cdot\text{h}\cdot\text{cm}^{-2}$) used up to now [15]. Second, the recent results obtained by geological comparison [3,4,6] are, in general, about 30% greater than the present semiempirical estimation; results by geological methods sometimes compare well with direct counting measurements, but these latter are, on the average, $\sim 20\%$ lower than the former ones. It is interesting to note that the earliest experimental determination of the half-life of ^{190}Pt performed by Hoffmann [8] using an ionization chamber is practically the same as the most recent half-life measurements by geological comparison by Cook *et al.* [4]. Good agreement with Graeffe's result [13] is also evidenced. Third, the uncertainties from different calculation models are, in general, larger than the ones associated with experimental determinations, especially in the case of geological comparison procedures. This may be caused by either oscillation in the calculation of the penetrability through the potential barrier, which contains the exponential Gamow's factor for decay given by the classical WKB-integral approximation [27], or uncertainties associated with parameter-values of some alpha-decay systematics. Finally, all sets of results from the different methods listed in Table 1 have furnished half-life values which, in some cases, differ from each other by a factor 2 or 3 (up to a factor 5 for values obtained by calculation models). Overall, the situation appears quite uncomfortable for the time being. The half-life value for the ^{190}Pt quoted in current Nuclide Tables is $(6.5 \pm 0.3) \times 10^{11}$ y, whereas a value of $(4.90 \pm 0.04) \times 10^{11}$ y is the one currently adopted to geochronological use. On the other hand, the quite good agreement observed between the present or the weighted average of calculated values ($(3.7 \pm 0.3) \times 10^{11}$ y or $(3.5 \pm 0.3) \times 10^{11}$ y, respectively) and the experimental weighted average from the counting experiments ($(3.9 \pm 0.2) \times 10^{11}$ y) suggests, at least provisionally, a value of $(3.9 \pm 0.3) \times 10^{11}$ y for the half-life of the ^{190}Pt isotope. However, precise knowledge of this quantity is still an unresolved issue, and this is expected to reflect on the accuracy of radioisotopic age determination of some ores and

meteorites. It is, therefore, required that researchers perform further experimental work, taking advantage of novel techniques developed for direct counting experiments, or carry out additional theoretical studies.

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Table 1 - Intercomparison between half-life measurements and calculated values for ^{190}Pt isotope*

Method	Author	Year	Experimental technique or type of model	Half-life (10^{11} y)	Ref.
Direct counting	Hoffmann	1921	Ionization chamber	~ 5	[8]
	Porschen and Riezler	1954	Nuclear-track emulsion	10	[9]
	Petrzhak and Yakunin	1961	Ionization chamber	4.7 ± 1.7	[10]
	Macfarlane and Khoman	1961	Ionization chamber	6.9 ± 0.5	[11]
	Graeffe and Nurmia	1961	Ionization chamber	6.8	[12]
	Graeffe	1963	Ionization chamber	5.4 ± 0.6	[13]
	Al-Bataina and Jänecke	1987	Ionization chamber	6.65 ± 0.28	[14]
	Tavares and Terranova	1997	CR-39 track detector	3.2 ± 0.1	[15]
Weighted average				3.9 ± 0.2	
Geological comparison	Walker <i>et al.</i>	1991	Pt-Os isochron for known age ore	8.8 ± 0.7	[1]
	Walker <i>et al.</i>	1997	Pt-Os isochrons for known age ores	4.49 ± 0.04^a	[2]
	Begemann <i>et al.</i>	2001	(revised value of Ref. [2])	4.69 ± 0.04^b	[6]
	Morgan <i>et al.</i>	2002	Pt-Os and Re-Os isochrons for sample ores	4.7 ± 0.3	[3]
	Cook <i>et al.</i>	2004	Pt-Os and Re-Os isochrons for IIAB iron meteorites	4.9 ± 0.1 $(5.1 \pm 0.1)^c$	[4]
			Pt-Os and Re-Os isochrons for IIIAB iron meteorites	4.90 ± 0.04 $(5.08 \pm 0.05)^c$	
Weighted average				4.78 ± 0.05	
Semiempirical calculation model	Taagepera and Nurmia	1961	two-parameter	2.4 ± 0.4 $(3.1 \pm 0.4)^d$	[16]
	Hornshøj <i>et al.</i>	1974	two-parameter	4.5 ± 0.7^e $(3.9 \pm 0.6)^d$	[18]
	Poenaru <i>et al.</i>	1986	six-parameter	12 ± 2^e	[19]
	Blendowske <i>et al.</i>	1991	one-parameter	2.8 ± 0.4^e	[20]
	Alex Brown	1992	three-parameter	8.7 ± 0.9^e	[21]
	Buck <i>et al.</i>	1993	four-parameter	3.8 ± 0.4^e	[22]
	Tavares <i>et al.</i>	1998	two-parameter	4.3 ± 0.2	[23]
	García <i>et al.</i>	2000	no-parameter	2.6 ± 0.3	[24]
	Dimarco <i>et al.</i>	2000	one-parameter	2.3 ± 0.3	[25]
	Tavares <i>et al.</i>	2005	one-parameter	3.7 ± 0.3	this work
Weighted average				3.5 ± 0.3	

* The presently adopted half-life-value is $(6.5 \pm 0.3) \times 10^{11}$ y [7].^a Assuming 0.0124% for the atomic percentage of ^{190}Pt .^b Assuming 0.01296% for the atomic percentage of ^{190}Pt .^c Adopting the value $(4.16 \pm 0.02) \times 10^{10}$ y for the half-life of ^{187}Re isotope [6].^d According to Ref. [17].^e Assuming a 10 keV uncertainty in the Q_α -value.

Table 2 - Input data for alpha decay systematics of platinum ($Z = 78$) isotopes^a.

Mass number A	Total half-life $T_{1/2}$ (y)	Branching ratio ^b b_α (%)	Q_α -value ^c (MeV)	Experimental, partial alpha decay half-life $T_{1/2_\alpha}^e$ (y)
166	0.950×10^{-11}	100	7.317	0.950×10^{-11}
167	2.22×10^{-11}	100	7.187	2.22×10^{-11}
168	0.633×10^{-10}	99.3	7.027	0.637×10^{-10}
170	4.37×10^{-10}	98	6.739	4.46×10^{-10}
171	1.39×10^{-9}	98	6.637	1.42×10^{-9}
172	3.12×10^{-9}	77	6.497	4.05×10^{-9}
174	2.82×10^{-8}	76	6.216	3.71×10^{-8}
175	0.798×10^{-7}	64	6.210	1.25×10^{-7}
176	2.01×10^{-7}	38	5.917	5.29×10^{-7}
177	3.35×10^{-7}	4.96	5.675	0.675×10^{-5}
178	0.668×10^{-6}	7.47	5.605	0.894×10^{-5}
180	1.65×10^{-6}	0.30	5.269	5.50×10^{-4}
181	1.65×10^{-6}	7.4×10^{-2}	5.182	2.23×10^{-3}
182	4.18×10^{-6}	3.8×10^{-2}	4.984	1.10×10^{-2}
183	1.24×10^{-5}	0.96×10^{-2}	4.855	1.29×10^{-1}
184	3.29×10^{-5}	1.7×10^{-3}	4.634	1.94×10^0
186	2.37×10^{-4}	1.4×10^{-4}	4.352	1.69×10^2
188	2.79×10^{-2}	2.6×10^{-5}	4.040	1.07×10^5
190 ^d	————	100	3.283	————

^a Data are taken from Refs. [7,28].

^b The quoted values are referred to ground-state to ground-state alpha transitions of $\ell = 0$.

^c This includes the screening effect due to the surrounding electrons.

^d The half-life-data for ^{190}Pt are those listed in Table 1 (direct counting experiments and geological comparison methods).

Figure Captions

Fig. 1 - Partial alpha decay half-life, $T_{1/2\alpha}$, plotted against $Q_\alpha^{-1/2}$ of ground-state to ground-state alpha transitions of $\ell = 0$ for ten isotopic sequences of alpha-emitter nuclides as indicated near the lines; full symbols represent experimental data from Refs. [7,28]; the lines are least-squares fitting to the calculated (open symbols) $T_{1/2\alpha}$ -values following the model detailed in [27]. Q_α is the Q -value for alpha decay corrected for the electron screening effect.

Fig. 2 - The same as in Fig. 1 for Pt isotopes. Full symbols are experimental data from Refs. [7,28] and Table 1. Open symbols represent calculated values according to our semiempirical model (see text). The inset evidences the case for ^{190}Pt isotope.

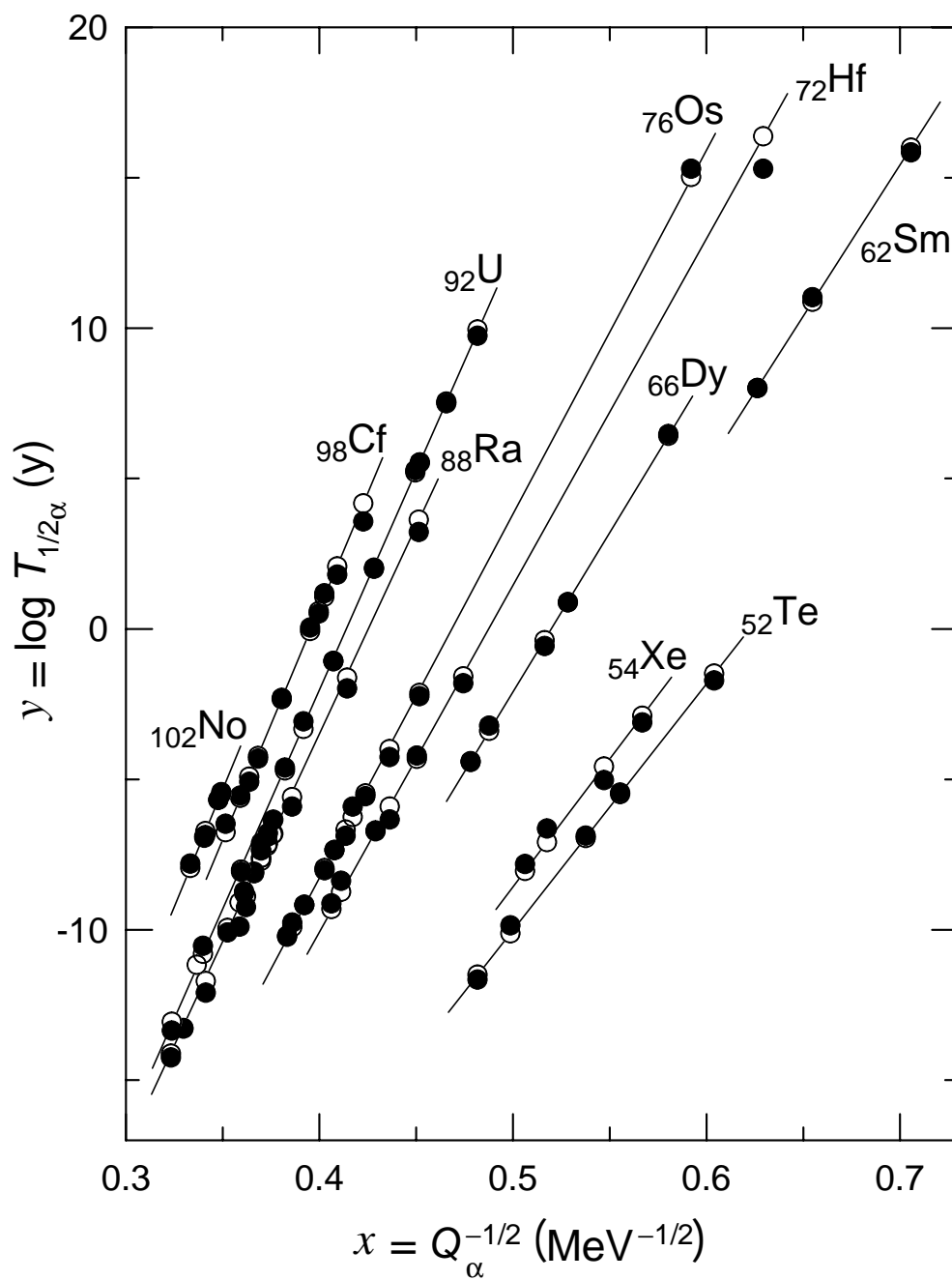


Fig. 1

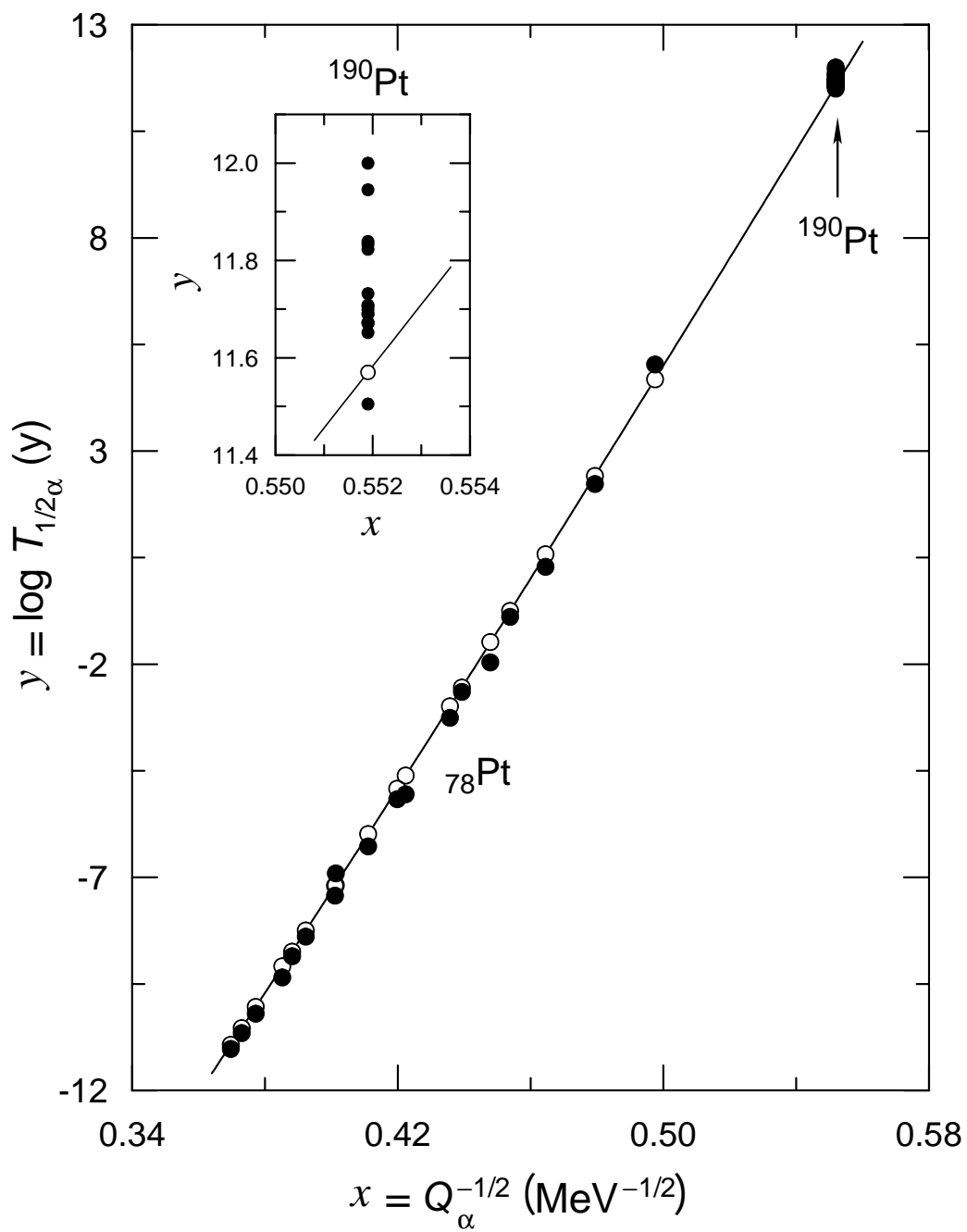


Fig. 2