

# The Discoverers of the Palladium Isotopes

THE THIRTY-FOUR KNOWN PALLADIUM ISOTOPES FOUND BETWEEN 1935 AND 1997

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*This is the fourth in a series of reviews of circumstances surrounding the discoveries of the isotopes of the six platinum group elements. The first review, on platinum isotopes, was published in this Journal in October 2000, the second, on iridium isotopes, was published here in October 2003 and the third, on osmium isotopes, was published in October 2004 (1). The current review looks at the discovery and the discoverers of the thirty-four isotopes of palladium.*

Of the thirty-four known isotopes of palladium, six occur naturally with the following authorised isotopic abundances (2):

The Naturally Occurring Isotopes of Palladium	
Mass number	Isotopic abundance, %
<sup>102</sup> Pd	1.02
<sup>104</sup> Pd	11.14
<sup>105</sup> Pd	22.33
<sup>106</sup> Pd	27.33
<sup>108</sup> Pd	26.46
<sup>110</sup> Pd	11.72

These naturally occurring isotopes were discovered by Arthur J. Dempster in 1935 (3) at the University of Chicago, Illinois, using a new mass spectrograph made to his design, although only the mass numbers were observed. The actual isotopic abundances were determined for the first time in the following year by Sampson and Bleakney (4).

## Artificial Palladium Isotopes

In 1935, using slow neutron bombardment, Amaldi *et al.* (5) identified two palladium activities with half-lives of 15 minutes and 12 hours. The latter value appeared to confirm a half-life of 14 hours that had been obtained earlier that year by McLennan, Grimmett and Reid (6). In 1937, Pool, Cook and Thornton (7) obtained similar half-lives of 18 minutes and 12.5 hours, and Kraus and Cork (8) were able to show experimentally, in that year, that these two activities belonged to <sup>111</sup>Pd and <sup>109</sup>Pd, respectively. Other slow neutron bombard-

ment activities found for palladium, such as a half-life of six hours discovered by Fermi *et al.* in 1934 (9) and half-lives of 3 minutes and 60 hours discovered by Kurchatov *et al.* (10) in 1935 do not appear to have been confirmed.

In 1940, Nishima *et al.* (11) obtained an unspecified activity with a half-life of 26 minutes which is also likely to have been <sup>111</sup>Pd. The actual half-life of <sup>111</sup>Pd is now known to be 23 minutes, so the different values obtained above are probably indicative of calibration problems.

These unspecified activities raise problems concerning the precedence for treating each discovery in this paper. Once the properties of an isotope are established and it is obvious that an unspecified activity must have been due to this particular isotope, then the activity is assigned to that isotope and can be regarded as being “the discovery”. However, using the definition that the primary criterion for discovery is the determination of both the atomic number and the mass number, these unspecified activities are included here only in the Notes to the Table that accompanies the Table of The Discoverers of the Palladium Isotopes.

Literature manuscript dates and conference report dates can be either the actual year of discovery or close to it, so when they are placed in the public domain these dates can be considered as being the “year of discovery”. However, complications arise with internal reports, especially if they represent the actual discovery, since they may not become publicly known until several years later. In these cases the historical date must obviously take precedence over the public domain date. As an



**Frédéric Joliot-Curie**  
1897–1956

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Born in Paris, Irène Curie was the daughter of scientists Pierre and Marie Curie, and therefore it was hardly surprising that she was academically brilliant. She became her mother's assistant at the Radium Institute, Paris, when only 21 years old and showed excellent aptitude in the use of the laboratory's instrumentation.

Frédéric Joliot was also born in Paris and in his twenties studied at the major Paris industrial engineering school, the *École Supérieure de Physique et de Chimie Industrielle*, under the tutelage of the physicist Paul Langevin, a friend of Marie Curie. Langevin suggested that Joliot should be considered for a post at the Radium Institute. Here Joliot met Irène Curie, who he married in 1926, adopting the surname Joliot-Curie.

After Frédéric had carried out major work to improve the sensitivity of the Wilson cloud chamber for detecting charged atomic particles, the Joliot-Curies became interested in the work of the German physicists Walther Bothe and Hans Becker who had noted that strong radiations were emitted when light elements were bombarded with alpha particles. The Joliot-Curies owned the major source of alpha particles available at that time – polonium which had accumulated over many years at the Radium Institute. They used this source to bombard aluminium foil, and found first neutron emission, followed by a long period of

**Irène Joliot-Curie**  
1900–1958



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positron radiation. They concluded that they had produced a new isotope of phosphorus of mass 30, compared to mass 31 found in natural phosphorus, and that the positron emission represented the decay of this isotope. This is the first example of the production of an artificial radioactive isotope. The discovery was announced in January 1934, and for this work they were awarded the 1935 Nobel Prize in Chemistry. Now, more than seventy years later, there are over 2700 artificial radioactive isotopes.

Like many physicists in the late 1930s the Joliot-Curies carried out research on nuclear fission that eventually led to both the atom bomb and nuclear power. After the war Frédéric convinced the French government to set up its own Atomic Energy Commission and he became its first High Commissioner. Irène succeeded her mother in becoming the Director of the Radium Institute. However, this was the time of the Cold War and Frédéric had strong left-wing political views. In 1950 Frédéric was dismissed from his post. Undaunted, both put great effort into helping to set up a large particle accelerator and laboratory complex at Orsay, south of Paris (*Institut de Physique Nucléaire d'Orsay*). This is now considered to be one of the major physics institutes in the world.

Irène died in 1956 and Frédéric in 1958, both from diseases related to prolonged exposure to radiation.

example, Brosi's discovery of  $^{103}\text{Pd}$  (12, 13) was given in an unpublished internal report dated July 1946 and was not mentioned publicly until its

inclusion in the 1948 "Table of Isotopes" (14). Therefore the discovery was not placed in the public domain until 1948, although 1946 is obviously

The Discoverers of the Palladium Isotopes						
Mass number	Half-life	Decay modes	Year of discovery	Discoverers	References	Notes
91	ps	EC + $\beta^+$ ?	1994	Rykaczewski <i>et al.</i>	17, 18	
92	1.1 s	EC + $\beta^+$	1994	Hencheck <i>et al.</i>	19	A
93	1.07 s	EC + $\beta^+$	1994	Hencheck <i>et al.</i>	19	B
94	9.0 s	EC + $\beta^+$	1982	Kurcewicz <i>et al.</i>	24	
95	–	EC + $\beta^+$ ?	–	–	–	C
95m	13.3 s	EC + $\beta^+$ , IT	1980	Noite and Hick	26	
96	2.03 m	EC + $\beta^+$	1980	Aras, Gallagher and Walters	27	
97	3.10 m	EC + $\beta^+$	1969	Aten and Kapteyn	28	
98	17.7 m	EC + $\beta^+$	1955	Aten and De Vries-Hamerling	29	D
99	21.4 m	EC + $\beta^+$	1955	Aten and De Vries-Hamerling	29	E
100	3.63 d	EC	1948	Lindner and Perlman	32	
101	8.47 h	EC + $\beta^+$	1948	Lindner and Perlman	32	
102	Stable	–	1935	Dempster	3	
103	16.991 d	EC	1946	1. Brosi 2. Matthews and Pool	12, 13 33	F
104	Stable	–	1935	Dempster	3	
105	Stable	–	1935	Dempster	3	
106	Stable	–	1935	Dempster	3	
107	$6.5 \times 10^6$ y	$\beta^-$	1949	Parker <i>et al.</i>	34	G
107m	21.3 s	IT	1957	Schindewolf	35	H
108	Stable	–	1935	Dempster	3	
109	13.7012 h	$\beta^-$	1937	Kraus and Cork	8	
109m	4.696 m	IT	1951	Kahn	38	I
110	Stable	–	1935	Dempster	3	
111	23.4 m	$\beta^-$	1937	Kraus and Cork	8	J
111m	5.5 h	IT, $\beta^-$	1952	McGinnis	40	
112	21.03 h	$\beta^-$	1940	Nishina <i>et al.</i>	11	
113	1.55 m	$\beta^-$	1953	Hicks and Gilbert	41	
113m	300 ms	IT	1993	Penttilä <i>et al.</i>	42	K
114	2.42 m	$\beta^-$	1958	Alexander, Schindewolf and Coryell	46	
115	25 s	$\beta^-$	1987	Fogelberg <i>et al.</i>	44, 45	
115m	50 s	$\beta^-$ , IT	1958	Alexander, Schindewolf and Coryell	46	L
116	11.8 s	$\beta^-$	1970	Aronsson, Ehn and Rydberg	47	
117	4.3 s	$\beta^-$	1968	Weiss, Elzie and Fresco	48	
117m	19.1 ms	IT	1989	Penttilä <i>et al.</i>	49, 50	
118	1.9 s	$\beta^-$	1969	Weiss <i>et al.</i>	51	
119	920 ms	$\beta^-$	1990	Penttilä <i>et al.</i>	50	
120	492 ms	$\beta^-$	1992	Janas <i>et al.</i>	52, 53	
121	285 ms	$\beta^-$ ?	1994	Bernas <i>et al.</i>	54	
122	175 ms	$\beta^-$ ?	1994	Bernas <i>et al.</i>	54	
123	174 ms	$\beta^-$ ?	1994	Bernas <i>et al.</i>	55	
124	38 ms	$\beta^-$ ?	1997	Bernas <i>et al.</i>	55	

ps = particle stable; IT = isomeric transition

## Notes to the Table

A	<sup>92</sup> Pd	Hencheck <i>et al.</i> (19) only determined the isotope to be particle stable. The half-life was first determined by Wefers <i>et al.</i> in 1999 (20).
B	<sup>93</sup> Pd	Hencheck <i>et al.</i> (19) only determined the isotope to be particle stable. The half-life was first accurately determined by Schmidt <i>et al.</i> (21) and Wefers <i>et al.</i> (22, 23) in 2000. A preliminary half-life of 9.3 s, determined by Wefers <i>et al.</i> in 1999 (20), was later withdrawn (22).
C	<sup>95</sup> Pd	The ground state has not been discovered, but Schmidt <i>et al.</i> (25) have suggested that the half-life is probably between 1.7 and 7.5 s from a consideration of the decay characteristics of <sup>95</sup> Rh.
D	<sup>98</sup> Pd	Aten and De Vries-Hamerling (30) first suggested the existence of this isotope in 1953. The discovery was independently confirmed by Katcoff and Abrash in 1956 (31).
E	<sup>99</sup> Pd	The discovery by Aten and De Vries-Hamerling (29) in 1955 was independently confirmed by Katcoff and Abrash in 1956 (31).
F	<sup>103</sup> Pd	The unpublished 1946 internal report of Brosi (12, 13) was not made public knowledge until included in the 1948 "Table of Isotopes" (14). Thus, Matthews and Pool appeared to be unaware of it in 1947 and their discovery can be considered to be independent.
G	<sup>107</sup> Pd	The unpublished 1949 internal report of Parkes <i>et al.</i> (34) was not made public until its inclusion in the 1953 "Table of Isotopes" (15).
H	<sup>107m</sup> Pd	Both Flammersfeld in 1952 (36) and Stribel in 1957 (37) observed this isotope but both ascribed it to <sup>105m</sup> Pd for which Schindewolf (35) could find no evidence.
I	<sup>109m</sup> Pd	The discovery by Kahn (38) was given in an unpublished 1951 report and was not made public until its inclusion in the 1953 "Table of Isotopes" (15). Flammersfeld (36) observed this isotope in 1952 but could not decide whether or not it was <sup>107m</sup> Pd or <sup>109m</sup> Pd.
J	<sup>111</sup> Pd	Segrè and Seaborg (39) appeared to dispute the discovery by Kraus and Cook (8) since their half-life of 26 m differed significantly from that of 17 m determined by the latter. However Kraus and Cook appear to have definitely identified the 180 h (7.45 d) half-life daughter isotope <sup>111</sup> Ag.
K	<sup>113m</sup> Pd	Meikrantz <i>et al.</i> (43) suggested the existence of an isomer of <sup>113</sup> Pd with a half-life exceeding 100 s; such an isomer should have been observed by Fogelberg <i>et al.</i> (44) but was not found.
L	<sup>115m</sup> Pd	According to Fogelberg <i>et al.</i> (45), Alexander, Schindewolf and Coryell (46), as with later observations of this isotope, may only have been observing a mixture of the ground state and isomeric state. Therefore the discovery of the pure isomeric state should probably be credited to Fogelberg <i>et al.</i> (44) in 1987.

## Some of the Terms Used for this Review

Atomic number	the number of protons in the nucleus
Mass number	the combined number of protons and neutrons in the nucleus
Nuclide and isotope	A nuclide is an entity characterised by the number of protons and neutrons in the nucleus. For nuclides of the same element the number of protons remains the same but the number of neutrons may vary. Such nuclides are known collectively as the isotopes of the element. Although the term isotope implies plurality it is sometimes used loosely in place of nuclide.
Half-life	the time taken for the activity of a radioactive nuclide to fall to half its previous value
Electron volt (eV)	The energy acquired by any charged particle carrying a unit (electronic) charge when it falls through a potential of one volt, equivalent to $1.602 \times 10^{-19}$ J. The more useful unit is the mega (million) electron volt, MeV.

Decay Modes	
$\alpha$	Alpha decay is the emittance of alpha particles, which are ${}^4\text{He}$ nuclei. Thus the atomic number of the daughter nuclide is lower by two and the mass number is lower by four.
$\beta^-$	Beta or electron decay for neutron-rich nuclides is the emittance of an electron (and an anti-neutrino) as a neutron decays to a proton. The mass number of the daughter nucleus remains the same but the atomic number increases by one.
$\beta^+$	Beta or positron decay for neutron-deficient nuclides is the emittance of a positron (and a neutrino) as a proton decays to a neutron. The mass number of the daughter nucleus remains the same but the atomic number decreases by one. However, this decay mode cannot occur unless the decay energy exceeds 1.022 MeV (twice the electron mass in energy units). Positron decay is always associated with orbital electron capture (EC).
EC	Orbital electron capture. The nucleus captures an extranuclear (orbital) electron which reacts with a proton to form a neutron and a neutrino, so that, as with positron decay, the mass number of the daughter nucleus remains the same but the atomic number decreases by one.
IT	Isomeric transition, in which a high energy state of a nuclide (isomeric state or isomer) usually decays by cascade emission of $\gamma$ (gamma) rays (the highest energy form of electromagnetic radiation) to lower energy levels until the ground state is reached. However, certain low-level states may also decay independently to other nuclides.

the proper historic year and must be treated as the actual “year of discovery”.

A technique is used for light and medium-heavy nuclides in which the nuclide fragments from a nuclear reaction are guided into a time-of-flight mass spectrometer.

The atomic numbers and the mass numbers of the detected nuclides can be determined by measuring: the total kinetic energy of the beam, the loss in energy when the beam is injected into an ionisation chamber and the actual time of flight. The determination of these numbers satisfies the criteria of discovery.

For detection in the mass spectrometer, the lifetimes of the nuclides must exceed 500 nanoseconds and must therefore be particle stable. It is expected that particle unstable nuclides, that is those that emit protons for the lighter isotopes of an element and those which emit neutrons for the heavier isotopes, are likely to have extremely short half-lives in this mass region. If, statistically, a nuclide should have been detected by the sensitivity of the technique, but was not, then it is likely to be particle unstable and thus can also satisfy the criteria of discovery, especially if it confirms theoretical predictions. None of the lighter and heavier palladium isotopes discovered by this technique have proved to be particle unstable, so the proton

and neutron drip lines have not yet been reached for this element.

Selected half-lives in the Table of ‘The Discoverers of the Palladium Isotopes’ are from the revised NUBASE database (16), except for those of masses 120 to 124 which are from the later measurements of Montes *et al.* (56). The criteria for discovery are generally those adopted in the review on platinum (1).

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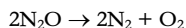


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## Nitrous Oxide Greenhouse Gas Abatement Catalyst

Among the naturally occurring greenhouse gases, nitrous oxide (N<sub>2</sub>O) is estimated to absorb 310 times (1, 2) more heat per molecule than carbon dioxide, thus contributing substantially to global warming (3). Atmospheric N<sub>2</sub>O is estimated to have increased by ~ 16% since the Industrial Revolution, and has contributed 6 to 11% to enhancing the greenhouse effect. Up to 40% of total atmospheric N<sub>2</sub>O is estimated to be man-made – equivalent to ~ 15 million tonnes per year (4). N<sub>2</sub>O is gradually accumulating in the atmosphere (2), despite slow breakdown by sunlight.

To reduce the production/emission of N<sub>2</sub>O as a waste product from nitric acid plants, the Norwegian nitrogen fertiliser manufacturer Yara International ASA (5) has developed a N<sub>2</sub>O abatement catalyst based on the reaction:



The de-N<sub>2</sub>O catalyst, which can cope with the high temperatures and corrosive environment of a nitric acid plant, is placed under the rhodium-platinum gauze pack and the catchment gauzes (6). It enables the N<sub>2</sub>O output in most plants to be reduced by 80% or more. The catalyst is of pelleted configuration, and when used with the Pt-Rh catalyst system gives an environmentally enhanced process with highly efficient N<sub>2</sub>O abatement. The catalyst is installed in several nitric acid plants, and more are planned.

Johnson Matthey, as a catalyst gauze supplier, will market the catalyst to 'clean development mechanism' (CDM) and 'joint implementation' (JI) countries as defined by the Kyoto Protocol. N<sub>2</sub>O emission reductions can thus be brought into line with requirements sought by the Kyoto Protocol (7). T. KOPPERUD

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