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# BETA-PARTICLE-ORBITAL ELECTRON CAPTURE BRANCHING AND "MISSING" BETA STABLE ISOTOPES

Geoffrey Wilkinson

February 14, 1948

Berkeley, California

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# Beta-Particle-Orbital Electron Capture Branching and "Missing" Beta Stable Isotopes

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## ABSTRACT

A consideration of hard negative electrons emitted by several radioactive isotopes of the heavier elements of odd atomic number and even mass number, has led to the conclusion that negative beta-particle-orbital electron capture branching occurs in these isotopes. This in turn has led to the postulation of unreported beta-stable isotopes of several elements of even atomic number.

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February 14, 1949

# Beta-Particle-Orbital Electron Capture Branching and "Missing" Beta Stable Isotopes

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The existence of "missing" stable, even Z, even A isotopes of the heavier elements has been discussed often by many in consideration of the stability of isotopes.[1] Such isotopes may be beta

(1) cf. T. P. Kohman, Phys. Rev. 73, 16 (1948).

stable and either of zero abundance, or present in nature in such low abundance as to be undetected by present techniques of mass spectrometry; that further isotopes may be found with improved techniques is shown by the recent discovery of $Dy^{156}$.[2] Since

(2) D. C. Hess, Jr., M. G. Inghram, Phys. Rev. 74, 1724 (1948).

elements above the middle of the periodic table are, on the average, energetically unstable to α-particle emission, some of the isotopes may be α-unstable with cosmologically short life times and may be missing in nature for this reason.[1,3] Indirect evidence has

(3) T. P. Kohman, US AEC Declassified Document AECD-2060.

now been obtained in this laboratory for the existence of several unreported beta stable isotopes.

A 15.4-day tantalum activity[4], now unquestionably allocated

(4) G. Wilkinson, H. G. Hicks, Phys. Rev. 74, 1733 (1948).

to mass 178 on the basis of the yield of the Lu-α-n reaction at various energies of bombarding α -particles, was found to decay almost entirely by negative beta-particle emission. The postulation of a beta stable tungsten isotope of mass 178 therefore followed. Additional evidence was obtained from bombardments of tantalum with 40, 50, 60 and 200 Mev deuterons from the 184-inch cyclotron; the presence of the 15.4 day $Ta^{178}$ was not detected in decays of the tantalum fractions and an upper limit of $10^{-3}$ can be set for the yield of this isotope in comparison with yields of $Ta^{177}$ and $Ta^{179}$.

A consideration of hard negative electron emission observed in other neutron deficient isotopes of odd Z, even A, decaying predominantly by orbital electron capture, has led to the conclusion that beta particle branching occurs in the following radioactive isotopes: $Tb^{156}$, $Ho^{162}$, $Ho^{164}$, $Tm^{166}$, $Tm^{168}$, $Lu^{174}$, $Ta^{178}$, $Ta^{180}$, $Re^{182}$, $Re^{124}$, $Ir^{190}$, $Au^{194}$, $Au^{196}$ and possibly $Tb^{154}$, $Lu^{170}$, $Lu^{172}$, $Ta^{176}$, $Au^{192}$. Several of the new radioactive isotopes have been briefly described,[4,5] while details of the radiation characteris-

(5) G. Wilkinson, Phys. Rev. 73, 252 (1948).

tics, mass allocation, etc. of some of these will be published shortly[6,7]; data on other new radioactive isotopes will be

(6) G. Wilkinson, "Radioactive Isotopes of Pt and Au", Phys. Rev. (April 1, 1949).
(7) G. Wilkinson, H. G. Hicks, "Radioactive Isotopes of the Rare Earth Isotopes, Part I. Thulium Isotopes", Phys. Rev. (May 1, 1949).

published subsequently. Beta particle branching might have been expected to occur in some of these isotopes, being, as they are, "shielded" by known stable isotopes; indeed, for $Au^{196}$ and $Re^{184}$, beta-particle emission has been previously recognized by other workers.[8] However, in the remaining cases, beta-particle branching also occurs, and the existence of the following beta stable isotopes must therefore be postulated - $Yb^{166}$, $W^{178}$, $Os^{182}$, $Pt^{190}$, $Hg^{194}$, and possibly $Dy^{154}$, $Hf^{170}$, $Hf^{172}$, $W^{176}$, and $Hg^{192}$. The evidence may be summarized as follows:

(8) G. T. Seaborg and I. Perlman, Rev. Mod. Phys. 20, 585 (1948).

(1) Radioactive isotopes of odd Z, even A, which could decay by negative beta-particle emission to known, or "missing" stable isotopes have been found to emit hard electrons whose aluminum absorption is that of a beta-particle, and whose energy distribution on a crude beta-ray spectrograph appears to be continuous with a well defined maximum energy.

(2) The energies of the hard electrons observed do not correspond with those to be expected from conversion of gamma radiation present. The conversion coefficient for hard gamma radiation to give the high electron energies observed would be very unusual indeed.

(3) No hard electrons have been observed in light radioactive isotopes of odd Z, odd A, heavier and lighter by one mass unit than those showing such hard electrons, although high intensities of hard gamma radiation were present in many cases.

(4) In general, the ratio of beta-particle to electron capture branching in elements where two or more "shielded" isotopes are known, e.g., holmium, or are possible, e.g., thulium, decreases considerably with decreasing mass number.

(5) The observed beta-particle emission in the nuclei where "missing" isotopes would be formed e.g., $Tm^{166}$, $Ta^{178}$, $Au^{194}$ is similar to that in nuclei where the stable daughter nucleus is known, e.g., to $Ho^{162}$, $Re^{184}$, $Au^{196}$, etc.

(6) The number of examples of beta branching in nuclei in analogous positions now observed, provides in itself reliable evidence for the present postulates.

It should be noted that soft beta-particles of energy less than ~0.3 Mev, would not be recognized as such with the present experimental techniques, and further, that beta-particle emitting isomers possibly of short half-life may exist in addition to the isotopes already found.

In the case of $Pt^{190}$, a lower limit for the half-life for decay by orbital electron capture was set at ~5000 years,[6] since no evidence of the d,3n reaction was observed in the bombardment of iridium with 19 Mev deuterons. From an examination of new data obtained in this laboratory[9] on the decay of the radioactive

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(9) T. C. Chu, private communication.

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isotope $Ir^{190}$ sketchily reported[10] previously, a beta-particle

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(10) L. J. Goodman, M. L. Pool, Phys. Rev. 71, 288 (1947).

of maximum energy 0.6 Mev was found in addition to conversion electrons, x-rays and gamma radiation. Beta-particle branching to the missing $Pt^{190}$ was therefore established.

In view of present data, it is not unreasonable to postulate the existence of beta-stable isotopes, $Gd^{150}$, $Pb^{202}$(11) and

(11) Experimental evidence from the work of Templeton, Perlman and Howland of this laboratory, shows a minimum half-life of about a hundred years for $Pb^{202}$.

possibly lighter, even Z, even A isotopes of elements now discussed; evidence for their existence may be obtained from beta-particle branching of the appropriate radioactive isotope. The 12-day $Tl^{202}$ isotope, which has been poorly investigated may well show beta-particle branching for example. It should be also pointed out that some of the "missing" nuclei here postulated approach or may be within the region of $\alpha$-particle instability; with increasing neutron deficit, instability both to $\alpha$-emission and to decay by orbital electron capture will increase. Some of the "missing" isotopes do indeed form possible $\alpha$ disintegration chains ending in known stable nuclei, e.g.,

$Hg^{194} \longrightarrow Pt^{190} \longrightarrow Os^{186}$ (1.59% abundance)

$Os^{182} \longrightarrow W^{178} \longrightarrow Hf^{172} \longrightarrow Yb^{166} \longrightarrow Er^{162}$ (0.1% abundance)

There seems also no reason to suppose that neutron excess beta stable nuclei, as yet unreported, do not exist, and that evidence for these may be obtained from orbital electron capture branching; it is, however, more difficult to make neutron excess isotopes than neutron deficient isotopes.

Detection of beta-stable, possibly α-active isotopes of the even Z elements is possible by either a) separation of light fractions by the usual methods of isotope separation, with detection by radioactivity measurement or by mass spectrometry - this method would be particularly feasible for tungsten, which has a volatile fluoride, or b) bombardments of odd Z elements with high energy protons or deuterons - the most profitable cases from the experimental point of view are those of tantalum, iridium, gold and europium. A long bombardment of iridium with 19 Mev deuterons is now being made to produce $Pt^{190}$ by the d, 3n reaction.

The author wishes to express his appreciation to Professor G. T. Seaborg and his colleagues for helpful discussions, and to Mr. H. G. Hicks for able assistance in experimental work on new radioactive isotopes.

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Feb. 14, 1949