The Radioactive Isotopes of Nickel and Their Assignments

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As a result of discrepancies in the reported methods of production and in the assignments of the radioactive isotopes of nickel the following reactions were investigated: Fe(α, n), Co(d, $2n$), Ni(d, p), Ni(n, γ), and Ni(n, $2n$). The relative yields of the known 2.6-hour and 36-hour periods from fast and slow neutrons on nickel were compared with other neutron reactions. The results obtained are in agreement with the assignment of the 2.6-hour period to Ni^{63} and the 36-hour period to Ni^{57}. The possible existence of a short period in nickel, reported as a result of an Fe(α, n) reaction, is not established since it would be masked by the 2.1-minute oxygen period formed from the carbon present in iron. In the attempted separation of the O^{15} activity it was observed that under the prevailing experimental conditions the rate of removal of oxygen from bombarded iron approached the rate of decay of O^{15}.

INTRODUCTION

THREE radioactive isotopes of nickel of periods 2.6 hours, 36 hours and 2 minutes have been reported. The 2.6-hour period was first produced by activation of nickel with slow neutrons,[1] and later by activation of zinc, copper, and nickel with fast neutrons.[2] The 2.6-hour period was also observed by other investigators after the bombardment of nickel with slow and fast neutrons and with deuterons.[3]

In addition a 36-hour period in nickel was obtained from the bombardment of iron with alpha-particles and nickel with fast neutrons.[3] However, the 36-hour activity was not produced by either a deuteron or slow neutron bombardment of nickel and so was assigned to Ni^{57} as shown in Fig. 1. In contradiction to these results, the 36-hour period was recently reported to be observed after slow neutron and deuteron bombardment of nickel.[4] The activity was consequently assigned to Ni^{59}.

A 2-minute period was also reported as a result of the bombardment of iron with alpha-particles and nickel with fast neutrons. This activity was assigned to Ni^{57}. Observations of the regularities in families of odd nuclei differing by alpha-particle units led to the expectation of periods of approximately 2 minutes, 30 hours, and several years in nickel.[5, 6] These periods were assigned to Ni^{57}, Ni^{59}, and Ni^{63}, respectively, leaving the 2.6-hour period to be assigned to Ni^{65}.

Because of these predictions and the discrepancies in the experimental data, a reinvestigation of the various nickel activities was thought justifiable.

THE 2.6-HOUR PERIOD

Hilger No. 5477 H.S. nickel metal was bombarded with 10-Mev deuterons. Spectroscopic analysis indicates the presence of 0.015 percent iron, 0.03 percent carbon, and traces of copper and oxygen. After bombardment the metal was dissolved, carriers of the above-mentioned impurities and of other elements commonly present in nickel were added, and separation of nickel was made with dimethylglyoxime. The nickel precipitate was redissolved and after another addition of carriers was again precipitated. This procedure was repeated several times until the filtrates from nickel showed no activity when measured with an immersion type G-M tube.

The purified nickel fraction was then measured with a Wulf electrometer and an ionization chamber filled with Freon under two atmospheres pressure. The decay curve shown in Fig. 2 is the simple 2.6-hour period which was followed through 18 half-lives to one-tenth of background. Since this period has also been produced

[1] J. Rotblat, Nature **136**, 515 (1935).
[2] F. A. Heyn, Physica **4**, 1224 (1937).
[3] J. J. Livingood and G. T. Seaborg, Phys. Rev. **53**, 756 (1938).
[4] R. L. Doran and W. J. Henderson, Phys. Rev. **60**, 411 (1941).
[5] G. R. Dickson, P. W. McDaniel, and E. J. Konopinski Phys. Rev. **57**, 351 (1940).
[6] G. R. Dickson and E. J. Konopinski, Phys. Rev. **58**, 949 (1940).

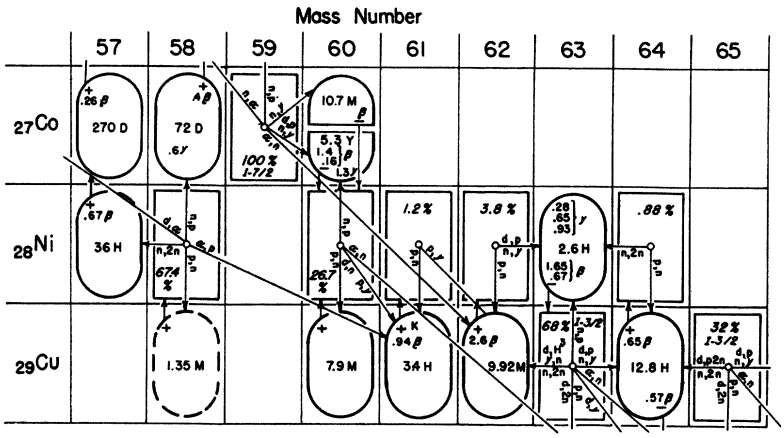

FIG. 1. Isotopic chart of the region studied.

by Zn(n, α) and Cu(n, p) reactions, its assignment to either Ni^{63} or Ni^{65} is possible. Definite assignment can best be based on the relative production of the activity by slow and fast neutrons on nickel.

In order to determine whether the apparent production by fast neutrons might actually be due to slow neutrons present in the fast neutron radiation, the yields of activity produced by slow and fast neutron bombardments were compared. These comparisons were made under the assumption that the capture cross section averaged over the slow neutron energies was nearly the same for the slow neutrons present in the paraffin as for the slow neutrons present in the fast neutron radiation.

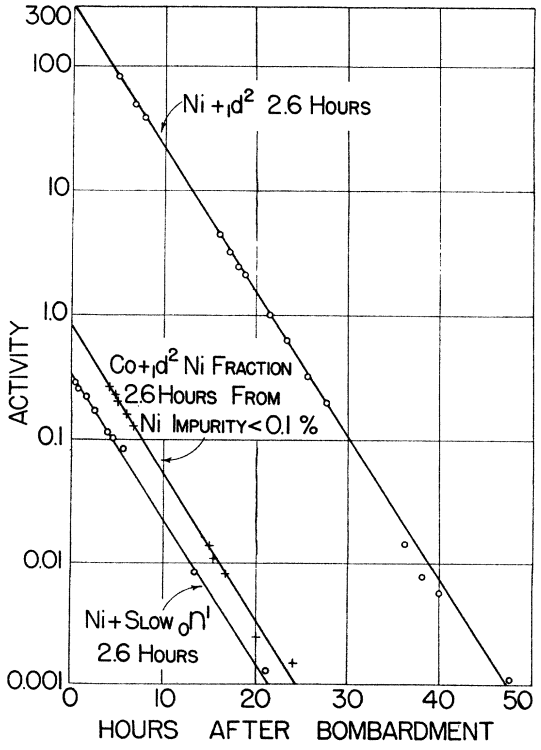

FIG. 2. Decay curves resulting from Ni+d, Co+d, and Ni+slow n bombardments.

Two similar sets of samples of manganese, silver, copper, and nickel were simultaneously bombarded with slow and fast neutrons. The samples to be activated by fast neutrons were placed directly behind a lithium target in the cyclotron tank (position A). The slow neutron samples were imbedded in paraffin blocks outside the tank (position B). The ratio of the yield obtained at position A to the yield obtained at position B for each element is given in Table I along with the type of reaction by which each of the activities was produced.

The ratio of the yield at A to the yield at B for the 2.6-hour nickel period is greater by a factor of 36 than that for the 2.59-hour manganese period which is produced by an (n, γ) reaction. However, the ratio is of the same order of magnitude as that for the 12.8-hour copper period which is produced by both (n, γ) and (n, $2n$) reactions. These comparisons indicate that the 2.6-hour nickel period is not produced by an (n, γ) reaction alone but by an (n, $2n$) reaction as well. This conclusion would necessitate the assignment of the activity to Ni^{63}.

THE 36-HOUR PERIOD

In the previously described bombardment of nickel with deuterons no 36-hour activity was found. If the 36-hour period had also been pro-

duced, its initial activity must have been less than 0.01 percent of that of the 2.6-hour period, otherwise it would have been detected. The ratio of the activities (36-hour : 2.6-hour) which would have been produced from an infinitely long bombardment was calculated to be less than 1 : 1200. In addition, it was found that during deuteron bombardments activities apparently due to deuterons were actually due to secondary neutrons produced at the target. Samples of nickel and cobalt were placed behind the primary nickel target. A weak activity was produced in the nickel sample, indicating the presence of the 2.6-hour period, and the known 2.59-hour period of Mn^{56} resulting from the $Co^{59}(n, \alpha)Mn^{56}$ reaction was observed in the cobalt sample. The intensity of the latter activity was about 1 percent of that produced by lithium neutrons.

Nickel was also bombarded with slow neutrons. No 36-hour activity was found but a 2.6-hour period was followed for seven half-lives to less than one-tenth of background. The ratio of the activities (36-hour : 2.6-hour) which would have been produced from an infinitely long bombardment was calculated to be less than 1 : 25.

To investigate a possible $Co(d, 2n)Ni$ reaction, Hilger No. 11045 V.P.S. cobalt metal was bombarded with deuterons. Spectroscopic analysis shows that its main impurities are nickel in a concentration less than 0.1 percent and traces of calcium, chromium, iron, copper, zinc, silver, and lead totaling about 0.1 percent. After bombardment, nickel was separated from iron, chromium, phosphorus, and other elements by precipitating the latter with ammonium hydroxide and filtering off the ammoniacal complex containing nickel. Since copper, cobalt, and other elements were still present, nickel was repeatedly separated from them with dimethylglyoxime as previously described. The decay curve of the nickel fraction again showed no 36-hour period but only the 2.6-hour period due to the nickel impurity. Thus no 36-hour activity was produced from a $Co(d, 2n)Ni$ reaction with 10-Mev deuterons.

TABLE I. Comparison of yields.

Reaction	Period	Yield at position A / Yield at position B
(n, γ)	2.59 hr. Mn	0.33
$(n, 2n)$	25 min. Ag	>5000
(n, γ) and $(n, 2n)$	12.8 hr. Cu	10
——	2.6 hr. Ni	12
——	36 hr. Ni	>200

The 36-hour period was produced by the known $Fe(\alpha, n)$ and $Ni(n, 2n)$ reactions, in which cases chemical separations similar to those previously described were made. However, the activity was not produced by $Co(d, 2n)$, $Ni(d, p)$ or $Ni(n, \gamma)$ reactions. All these data can be interpreted by assigning this period to Ni^{57}. The absence of activity from a $Co(d, 2n)$ reaction is consistent with the recently reported non-existence of stable Co^{57}.[7]

THE 2-MINUTE PERIOD

The 2-minute period has been previously reported as a result of $Fe(\alpha, n)Ni$, and $Ni(n, 2n)Ni$ reactions. However, the definite possibility of the presence of the 2.1-minute oxygen period which might complicate the identification of the nickel was not indicated. The 2.1-minute oxygen should always be formed by the $C^{12}(\alpha, n)O^{15}$ reaction after activation of iron with alpha-particles since even the purest iron samples contain some carbon. Therefore, an attempt was made to remove oxygen in order to observe the possible existence of a short period in nickel.

An independent alpha-particle bombardment of carbon in the form of sodium carbonate was made. After dissolving the activated substance in water a part was measured immediately with an immersion type G-M tube. Another part was treated with hydrochloric acid and evaporated. The procedure was repeated three times before measurement. The presence of a 2.2 ± 0.1-minute activity in both parts showed that the radioactive O^{15} was not entirely removed. The difficulty lies in the fact that the removal of oxygen is never complete in the procedure mentioned and that the rate of removal approached the rate of decay of the oxygen period.

Iron was bombarded with alpha-particles and then dissolved in nitric acid. The activity in one part was measured immediately and was found to decay with a 2.2-minute period. Another part of the same solution was treated with hydrochloric acid and evaporated. The solid salt of

[7] J. J. Mitchell, H. S. Brown, and R. D. Fowler, Phys. Rev. **60**, 359 (1941).

iron was converted into the oxide by ignition. The oxide was again dissolved and the operation repeated. Measurements on the G-M counter showed that a weak 2.2-minute period remained just as in the case of the carbon bombardment.

Nickel was also bombarded with fast neutrons, but no 2-minute period was observed. However, the production of such a period by fast neutron bombardment of oxygen in the form of sodium carbonate was confirmed.

These results indicate that the 2-minute activity reported as a result of the bombardment of iron with alpha-particles may be due to O^{15}. However, the results do not necessarily exclude the possibility of the existence of a short period in nickel, the presence of which is not evident from the above described experiments. The development of an experimental technique for the rapid removal of oxygen is necessary before the question of the existence of a short period in nickel can be settled.

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Forbidden Transitions in β-decay and Orbital Electron Capture and Spins of Nuclei

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A general formula giving minimum lifetimes for forbidden β-transitions of arbitrary order or forbidden orbital electron capture is derived. Exact Coulomb wave functions are used for the electron. It is shown that the observed electron emission of K^{40} requires Gamow-Teller selection rules. Combined with the Konopinski-Uhlenbeck result that only the tensor and vector interactions are compatible with the energy spectra of the β-rays from Na^{24}, P^{32}, and RaE, it follows that the tensor interaction alone can explain both the lifetimes and energy spectra of forbidden β-transitions. The application of the tensor interaction to K^{40} and to the other long-lived β-emitters, Rb^{87}, Lu^{176}, Be^{10}, C^{14}, and to existing data on orbital electron capture, leads to certain spin and parity predictions about parent and product nuclei—e.g., neither Be^{10} nor C^{14} can have a spin greater than $3\hbar$, the 2-Mev γ-ray from K^{40} is associated with K-electron capture to an excited state of A^{40} having even parity, etc. The stability of the known neighboring isobars and the conditions under which L-electron capture becomes more probable than K-electron capture are also discussed.

I. INTRODUCTION

EXPERIMENTS on β-radioactive nuclei have tended in recent years to confirm Gamow and Teller's[1] modification of Fermi's theory of β-decay according to which an "allowed" transition may involve a spin change of as much as one unit of $\hbar$. In particular, the strikingly large probability of the reaction $He^6 \rightarrow Li^6 + e^-$, was presumed to provide strong support for the correctness of the Gamow-Teller selection rules since the transition from He^6 with a spin zero to Li^6 with a spin one involves a spin change of one unit. The Li^6 spin has been measured but the spin of He^6 was based on the argument from nuclear theory that all nuclei, whether stable or unstable, with an even number of protons and an even number of neutrons, have spin zero. It is certainly true that all known *stable* even-even nuclei have spin zero but the extrapolation to unstable even-even nuclei has been rendered extremely dubious by the fact that certain homologues of He^6, such as Be^{10} and C^{14}, decay very slowly and cannot therefore have the same

[1] G. Gamow and E. Teller, Phys. Rev. **49**, 895 (1936); there is a good deal of indirect evidence for the Gamow-Teller selection rules [cf. especially E. P. Wigner, Phys. Rev. **56**, 519 (1939); and White, Creutz, Delsasso, and Wilson, Phys. Rev. **59**, 63 (1941)] but none can be as conclusive as the type discussed below.