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# A New Measurement of the Packing Fractions of the Nickel Isotopes

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The packing fractions of the nickel isotopes have been measured by comparison with known hydrocarbon lines using a double-focusing mass spectrograph. A new constant source of positive ions was used which produces mass spectra as high as the eighth order. The values of the packing fractions were determined to be: $^{58}$Ni($-8.29\times10^{-4}$); $^{60}$Ni($-8.69\times10^{-4}$); $^{61}$Ni($-8.75\times10^{-4}$); $^{62}$Ni($-9.01\times10^{-4}$); $^{64}$Ni($-8.62\times10^{-4}$). The binding energies of the neutrons added to form the heavier isotopes are tabulated. An explanation is suggested for the differences in the packing fractions reported here and those given by Okuda and his associates.

## INTRODUCTION

THE first determination of the packing fraction of nickel was made by Aston,[1] who used the doublets given by the approximate ratios $^{58}$Ni:CO$_2$ and CH$_4$:$^{12}$C. These measurements led to a value of $-10\times10^{-4}$ MU for the packing fraction of $^{58}$Ni. In 1938 Aston repeated his measurements[2] using his new spectrograph and the doublets $^{58}$Ni:CO$_2$ and $^{16}$O:$^{12}$C. These measurements gave a value of $-8.35\times10^{-4}$ MU for $^{58}$Ni. In these early measurements the source of ions was a high voltage discharge through a vapor of Ni(CO)$_4$, modified by the introduction of CO$_2$ to reduce the tendency of the discharge to decompose the Ni(CO)$_4$. In other experiments, the Ni(CO)$_4$ was cooled to $-20$°C and the pure vapor was passed continuously into the discharge tube. Notwithstanding the various precautions that were taken, the discharge was unsteady and spectra beyond the first order were not sufficiently intense to be useful.

In 1941 a new determination of the packing fractions of the nickel isotopes was reported.[3] The source of positive ions for this measurement was a high voltage discharge through a vapor mixture of Ni(CO)$_4$ and normal heptane, $n-$C$_7$H$_{16}$. No further details were given on the source but in a previous paper,[4] it was stated that a cylindrical discharge tube was used with an aluminum bar anode and a concave iron cathode. In the absence of more details it is difficult to judge the constancy or reproducibility of this source of positive ions.

Generally speaking, a high voltage gaseous discharge tends to produce an intermittent and fluctuating beam of ions. This occurs in many cases because of the tendency of the discharge to decompose the chemical compounds that are introduced and because of the accumulation of sputtered material. In view of the uncertainties of this type of source and the apparent rapid and irregular variation in the binding energy of the nickel isotopes, as deduced from the packing fraction measurements of Okuda[4] and his associates, it was deemed desirable to make new measurements under completely different conditions.

## SOURCE

The positive ions of nickel were derived from a new ion source,[5] the schematic representation of which is shown in Fig. 1. The tungsten crucible, which holds the metal to be vaporized, was made by drilling a hole 2.5-mm diameter, and 5-mm deep in a pure tungsten rod 5-mm diameter and 9.5-mm long. A flat spot was ground on the opposite end of this tungsten rod and a one-mm support rod of pure tungsten spotwelded to this point. The material to be vaporized was introduced into the crucible after which a tight fitting, pure tungsten cap was placed over the hole in the crucible. This cap as provided with an axial hole 0.25-mm diameter which limited the cross section of the beam of emerging vapor.

The crucible thus prepared was supported in the axis of a pure tungsten filament in the form of a circular loop. The filament was made of wire 0.425-mm diameter and operated at approximately 15 amperes. The plane of this filament was located 1.5-mm back from the plane of the tungsten cap. In this way electrons from the filament performed the double function of vaporizing the material within

TABLE I. Packing fractions $f_1$ of nickel isotopes.

| Doublet | Number of doublets | $(f_2-f_1)$ $\times10^4$ MU | $f_1\times10^4$ MU |
|---|---|---|---|
| $^{58}$Ni$^{+2}-^{12}$C$_2$H$_5$ | 7 | 24.96 | $-8.29\pm0.07$ |
| $^{60}$Ni$-^{12}$C$_5$ | 10 | 11.90 | $-8.69\pm0.08$ |
| $^{61}$Ni$-^{12}$C$_5$H | 9 | 13.24 | $-8.75\pm0.08$ |
| $^{62}$Ni$-^{12}$C$_5$H$_2$ | 10 | 14.74 | $-9.01\pm0.05$ |
| $^{64}$Ni$-^{12}$C$_4$$^{13}$CH$_3$ | 7 | 16.02 | $-8.62\pm0.08$ |

Note: The masses of $^{12}$C and H used in the calculations of the packing fractions $f_2$, and the mass of the neutron used in the calculation of the binding energies are values recommended by K. T. Bainbridge, *Isotopic Weights of the Fundamental Isotopes*, Preliminary Report No. 1, National Research Council, June 1948, p. 19. The mass of $^{13}$C was that given by S. Flügge-J. Mattauch, Physik. Zeits. **44**, 192 (1943).

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[1] F. W. Aston, Nature **120**, 956 (1927); cf. also *Mass Spectra and Isotopes* (Edward Arnold & Co., London, 1933).

[2] F. W. Aston, Nature **141**, 1096 (1938).

[3] T. Okuda, K. Ogata, H. Kuroda, S. Shima, and S. Shindo, Phys. Rev. **59**, 104 (1941).

[4] T. Okuda, K. Ogata, K. Aoki, and Y. Sugawara, Phys. Rev. **58**, 578 (1940).

[5] A. E. Shaw, Phys. Rev. **73**, 1222 (1948).

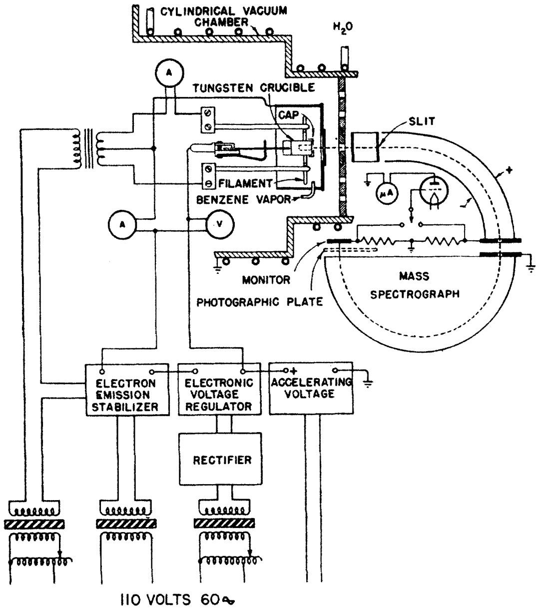

FIG. 1. Schematic diagram of positive ion source.

the crucible and ionizing it as it emerged through the hole in the cap. *It was found that this source produces spectra as high as the eighth order when the bombarding voltage is approximately 600.*

The thermal radiation given off by the hot crucible upsets the equilibrium of the filament by raising its temperature beyond the point corresponding to the power dissipated in it. Although the filament power may be controlled manually, for the best stability and reproducibility it is desirable to employ electronic regulating and stabilizing circuits. These are shown in block diagram in Fig. 1. Complete details of these circuits and the source will be published elsewhere.

The supply voltages for the source enter through 30-kv isolation transformers since the source proper is above ground by the magnitude of the accelerating voltage. The coarse controls are provided by the variacs shown, whereas the fine controls are attached to the individual emission stabilizer and voltage regulator. The product of the electron current to the crucible and the bombarding voltage is taken as a measure of the power dissipated in the crucible. This is only approximate but it serves as a convenient means for reproducing a fixed set of conditions.

## EXPERIMENTS

In order to utilize this source for packing fraction measurements of nickel, it was necessary to provide a standard hydrocarbon comparison line for each of the nickel isotope lines. This was accomplished by introducing benzene vapor ($C_6H_6$) directly into the ionization chamber through a small tantalum tube (see Fig. 1). Thus both the nickel vapor from the hot crucible and the benzene vapor were ionized simultaneously by the same 600-volt electrons. The hydrocarbon lines were sufficiently intense with pressure of the order of $5\times10^{-5}$ mm Hg. The nickel lines were brought out with comparable intensity by 30 watts of electron power dissipated in the crucible. The photographs were made with a double-focusing mass spectrograph,[6] in which the principal slit was set at 0.020 mm. Exposure times varied from three minutes to five minutes. The monitor shown in Fig. 1 provided a check of the intensity and constancy of the ion beam. When desired, the photographic plate is moved into the path of the beam by means of an externally controlled Wilson Seal.

## RESULTS

Second-order spectra were photographed to obtain a comparison line for $^{58}$Ni, since no ion corresponding to $m/e=58$ has been observed with benzene. First order spectra contained the four doublets which were used for the measurements of $^{60}$Ni, $^{61}$Ni, $^{62}$Ni, and $^{64}$Ni. The hydrocarbon comparison line for $^{64}$Ni was the $^{13}$C isotope peak.[7] In Fig. 2 is shown a photographic enlargement of a typical triplet from which the doublet $^{58}\mathrm{Ni}^{+2}-{}^{12}\mathrm{C}_2\mathrm{H}_5$ was obtained. Figure 2 also shows the four doublets in the first order.

All measurements of doublet separation were made on a Gaertner Comparator whose scale read directly in thousandths of a millimeter. Each final value of doublet separation was the result of five separate measurements that were generally reproducible to within ±0.002 mm. All measured values were given equal weight and expressed in terms of the probable error in the arithmetic mean. The packing fractions are given in Table I.

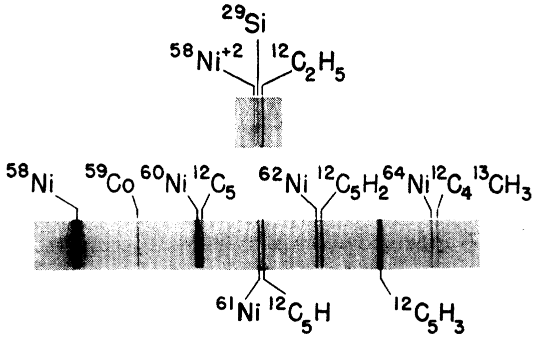

FIG. 2. Typical doublets used in calculation of packing fractions of nickel isotopes.

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[6] A. J. Dempster, Phys. Rev. **53**, 64 (1938).

[7] "Mass Spectral Data," National Bureau of Standards, Washington, D. C., Serial No. 175, June 30, 1948.

TABLE II. Binding energies of $n$ added neutrons.

| Mass No. A | Atomic mass $M$ | $\Delta M$ | $n$ | Binding energy | |
|---|---|---|---|---|---|
| | | | | Mass units | Mev |
| 58 | 57.9519 | | | | |
| | | 1.9960 | 2 | 0.0218 | 20.3 |
| 60 | 59.9479 | | | | |
| | | 0.9987 | 1 | 0.0102 | 9.5 |
| 61 | 60.9466 | | | | |
| | | 0.9975 | 1 | 0.0114 | 10.6 |
| 62 | 61.9441 | | | | |
| | | 2.0007 | 2 | 0.0171 | 16.0 |
| 64 | 63.9448 | | | | |

## DISCUSSION OF RESULTS

The packing fractions in Table I differ in several respects from those obtained by Okuda[3] and his associates, whose results were as follows:

$^{58}$Ni($f_1=-6.97\times10^{-4}$), $^{60}$Ni($f_1=-8.37\times10^{-4}$), $^{61}$Ni($f_1=-7.5\times10^{-4}$), $^{62}$Ni($f_1=-8.14\times10^{-4}$) and

$$^{64}\mathrm{Ni}(f_1=-8.22\times10^{-4}).$$

The largest discrepancy between their results and the values in Table I occurs at $^{58}$Ni. The source of ions for their measurements was a high voltage discharge through a vapor mixture of Ni(CO)$_4$, and normal heptane, $n-\mathrm{C_7H_{16}}$, the latter providing the standard hydrocarbon comparison lines. The $^{58}$Ni mass was compared with a hydrocarbon mass that was assumed to be $^{12}\mathrm{C_4H_{10}}$. In the case of many hydrocarbon compounds, the ion whose $m/e=58$, occurs as a peak with the isotope $^{13}$C, for ionizing electrons of 50 and 70 volts. However, there are also numerous hydrocarbons in gaseous form such as 1, 2-diisopropylbenzene, $n$-butyl mercaptan and 2, 2-dimethylbutane whose mass spectra reveal the existence of normal peaks at $m/e=58$. In the case[8] of $n-\mathrm{C_7H_{16}}$, the ion formed by electron impact corresponding to $m/e=58$ is $^{12}\mathrm{C_3}^{13}\mathrm{CH_9}^+$ and not $^{12}\mathrm{C_4H_{10}}^+$, although it is conceivable this latter ion may occur in the high voltage discharge through the vapor mixture used.

If one compares the $^{58}$Ni mass with the mass $^{12}\mathrm{C_3}^{13}\mathrm{CH_9}$, their value of $f_1$ is increased to $-7.72\times10^{-4}$. In the case of $^{60}$Ni, Okuda and his associates used a comparison mass which was assumed to be $^{12}\mathrm{C_5}^+$. There is no evidence for the formation of this ion by electron impact,[8] although it may occur under the conditions of the high voltage discharge through the vapor mixture of Ni(CO)$_4$ and $n-\mathrm{C_7H_{16}}$. The hydrocarbon ions corresponding to $m/e=61$, 62, and 64 do occur normally[8] under impact of 50 and 70-volt electrons.

The values of the packing fractions deduced by Okuda and his associates are all slightly smaller than those obtained in the present observations. The differences may be due to the fact that in their experiments as in all previous observations, except for recent experiments reported by Dempster,[9] an intermittent gas discharge was used as a source of ions.

The packing fractions in Table I enable one to calculate the true atomic mass of each of the nickel isotopes. Each of these isotopes is formed from the preceding one by the addition of neutrons, hence the binding energy of the added neutrons can be calculated from the actual increment in mass. Table II gives the values of binding energy for the $n$ added neutrons in terms of mass units and Mev. The mass increase $\Delta M$ is in each case less than the neutron mass. Table II also reveals that the two neutrons involved in the formation of $^{60}$Ni from $^{58}$Ni are most firmly bound, while the additional pair added to $^{62}$Ni to form $^{64}$Ni are least firmly bound.

The atomic masses given in Table II enable one to calculate the atomic weight of the element nickel. Combining these masses and the relative abundances[10] of the nickel isotopes the atomic weight is found to be 58.70, referred to the chemical scale.

## ACKNOWLEDGMENT

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[8] "Mass Spectral Data," National Bureau of Standards, Washington, D.C., Serial No. 14, October 31, 1947.

[9] A. J. Dempster, Phys. Rev. **74**, 1225 (1948).

[10] J. R. White and A. E. Cameron, Phys. Rev. **74**, 991 (1948).

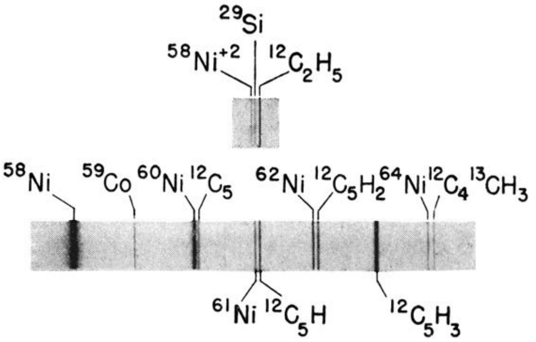

FIG. 2. Typical doublets used in calculation of packing fractions of nickel isotopes.