AN ATTEMPT TO ADD AN ELECTRON TO THE NUCLEUS OF AN ATOM

BY WILLIAM D. HARKINS AND WEBSTER B. KAY

ABSTRACT

Since the present theories of the atom indicate that an electron is attracted by the atom nucleus, they also suggest that the principal difficulty in accomplishing the addition of an electron is in obtaining such (unknown) conditions as make the attraction effective, Electrons with a velocity corresponding to 138,000 to 145,000 volts were caused to strike the surface of liquid mercury. The mercury served as an anti-cathode in an x-ray tube. (This work was done before the electron tube of Coolidge was available.) When the experiments were begun it was not expected that positive results would be attained, since either the voltage may be too low, or the number of electrons shot into the surface (4×10^{21}) too small, but it was thought to be of value to show this to be true. The addition of an electron to the nucleus of an atom of mercury should give an atom of gold. Since no gold was found in the exceedingly sensitive tests used, it is shown that either less than one in a billion of the electrons attached itself to an atom nucleus, or else all or a part of the nuclei produced were not sufficiently stable to endure for the period (24 to 48 hours) of the tests.

1. INTRODUCTION

'HE nucleus of an atom is supposed to have a netpositive chargeequal THE nucleus of an atom is supposed to have a heater of Z of the element. It has been
(in electron units) to the atomic number (Z) of the element. It has been found that an alpha-particle with a net positive charge of $2e$ may attach itself to the nucleus of an atom of nitrogen provided its velocity immediately before impact is about 1.8×10^9 cm per second (or about 7×10^6 electron equivalent volts of energy). One photograph obtained by Harkins and Shadduck indicates that such an attachment may occur at a velocity as low as 1.57×10^9 cm per second $(5.1 \times 10^6$ electron volts).

The scattering of alpha-particles by gold indicates that the inverse square law holds at distances between the centers of the nucleus of the gold square law holds at distances between the centers of the nucleus of the gold
atoms and that of the alpha-particle greater than 3.2×10^{-12} cm. Since the magnitude of the net positive charge of the nucleus of an atom of mercury is almost the same as that of gold, it may be considered that the above value is as valid for mercury as for gold. Now, if the inverse square law holds for is as valid for mercury as for gold. Now, if the inverse square law holds for
electrons also at distances of nuclear approach greater than 3.2×10^{-12} cm an electron should gain a velocity of about 0.991 c (2.9 \times 10¹⁰ cm/sec.) if an electron should gain a velocity of about 0.991 c (2.9 $\times10^{10}$ cm/sec.) i:
it falls from rest outside the atom to a distance of 3.2 $\times10^{-12}$ cm from the nucleus of the atom. This corresponds to the velocity given to an electron by a potential fall of three million volts.

Thus it seems that the electron should, provided it attains a condition; such that the net attractive effect of the nucleus becomes available, approach the nucleus of the atom of mercury with about 20 times the veIocity and more than one-half the kinetic energy of an alpha-particle which attaches. itself to the nucleus of a light atom.

If the above point of view is accepted, an interesting conclusion emerges. By present methods it is not possible to accelerate electrons by potentials greater than about 300,000 volts, except by the use of several Coolidge electron tubes in series. Such a potential is very small compared with the potential drop in the atom itself. Thus the use of the external electric field for the acceleration of electrons is needed only to drive the electron into the atom with sufficient (possibly critical) velocity and energy so that the attractive force between the nucleus and the electron may be effective. The magnitude of the energy essential for this purpose is unknown, but it may not be unreasonable to suppose that it is greater than that necessary to excite the hardest x-rays which the substance emits.

2. ATTEMPT TO ADD AN ELECTRON TO THE NUCLEUS OF AN ATOM OF MERCURY

In the experiments reported in the present paper a mercury surface was bombarded by electrons with an energy of 145,000 volts, which corresponds to a velocity of 1.9×10^9 cm/sec. While this voltage is much greater than that required to excite the hardest x-rays of mercury, it was the opinion of the writers that a much higher voltage would give a considerably greater chance of success in the addition of an electron to the nucleus of the mercury atom, but the voltage used was at the time the highest available for this work. On this account it was our wish to use an element of considerably lower atomic number. However, mercury¹ was selected for the work because (1) it may be separated almost entirely from the next lower element (gold), and (2) an exceedingly small amount of gold $(10^{-9}g)$ may be detected in a comparatively large amount of mercury (130g). It happened that we also possessed a considerable amount of mercury which is not only free from detectable amounts of gold, but is also considerably lighter than ordinary mercury (about 0.¹ unit). Now Aston had reported the presence in mercury of an isotope isobaric with gold (atomic weight=197). If this had been correct, our sample, produced by Harkins, Mulliken and Mortimer, would have been richer with respect to this isotope than any other known sample of mercury.

Later Aston found this observation to be erroneous, so the chance of producing gold would seem to be best with the mercury isotope of mass

¹ We were especially desirous of avoiding the use of mercury on account of the considerable notoriety attached to the finding of gold in mercury arc lamps by Meithe and Stammreich, (Miethe and Stammreich, Naturwissenschaften 12, 597, 744, 1211 (1924), 13, 635 (1925); Zeits. f. Anorg. Chem., 140, 368 (1924); 148, 93 149, 263 (1925); 150, 350 (1926); Phys. Zeits. 2&, 842 (1925)) and the idea that it was obtained by a transmutation of some of the mercury. In this work the total applied potential was only 220 volts, and the effective velocity of the electrons ejected corresponds to only a small fraction of the potential. Miethe's method has been tested by a number of investigators, (Haber, Jaenicke and Matthias, Zeits. f. Anorg. Chem. 513, 153 (1926); Piutti and Boggio-Lera, Giornale de Chemica industriale 8, 59. Rend. R. Accad. Napoli (S) 31, 194 (1926); Riesenfeld and Haase, Ber. der Deut. Chem. Ges. , 59, 1625 (1926); Tiede, Schleede, Goldschmidt-ibid. 1629; Sheldon, Estey, and Maily, Scientific American, 1925, p. 291), all of whom found that no gold was produced. Notices of the results of our own work were published two years ago (Metallbörse 16, 1117 (May 19, 1926), report of the results of the work of the writers) without our knowledge.

number 199. It is not unlikely, even if gold happened to be formed in our experiments, that the gold atoms produced were not sufficiently stable to exist until the chemical test was completed.

The only work on mercury in which a high total potential was used is that of Nagoaka.² He obtained a high potential discharge from an induction coil between a metal point in oil and a mercury surface. The velocity of the electrons which struck the surface of the mercury was not determined, but was probably only of the order of a few hundred or a few thousand volts, since the discharge was produced in oil at atmospheric pressure. However, Nagoaka found gold in the mercury after this treatment.

General method and the stability of atom nuclei produced. The general method used in this work was to accelerate the electrons from a Coolidge cathode in an x-ray tube in which 9 cc of mercury was used as an anticathode. The potential on the tube was 138,000 to 145,000 volts. The mercury was tested for gold after enough current had been passed to form 1.⁵ g of gold provided each electron should attach itself to the nucleus of a mercury atom. Since the limit of detection was about a billionth of a gram of gold, or slightly less, it was necessary for the detection of this element that the yield should be more than one trillion of its atoms, with a sufficient stability to have a life of from 24 to 48 hours, that is, the period of the test.

Apparatus and method. The method employed to bombard the mercury atoms with electrons involved the use of the mercury as a target for cathode rays of high velocity. This amounted to an x-ray tube with a mercury anticathode. Such a tube involves considerable change in the design of the anticathode on account of the high vapor pressure of mercury, which is 0.0004 millimeter at 0° C. Since the vacuum necessary for the successful operation of the tube is of the order of 10^{-6} millimeter pressure, it is evident that even at O'C this vapor pressure is much too great. This difficulty is increased by the fact that the heating effect at the anticathode by the cathode rays makes the vapor pressure still greater. It is obvious, therefore, that the mercury must be kept at a temperature considerably below 0° C. Extrapolation of the empirical equation for the vapor pressure of mercury showed that at about -30° C its vapor pressure would be sufficiently low. Of the various cooling agents available with which to obtain a temperature in this neighborhood, liquid ammonia was the only low boiling liquid available in sufficient quantities.

The construction of the tube is shown in Fig. 1. The principal parts are the discharge chamber A , the cathode B , and the anticathode C . The discharge chamber is made from a one liter round bottom Pyrex flask. The Coolidge cathode is a medium focus spiral filament of tungsten which is heated by a 6 volt storage battery. The anticathode resembles an inverted Dewar bottle with the outer jacket fused to the bottom of the flask. A shallow cup in which the mercury is held is made in the end of the inner

² Nagoaka, Naturwiss, 13, 684 (1925), 14, 85 (1926), Nature 116, 95 (1925). J. de phys. et de Radium 6, 209 (1925); Soc. Franc. de Phys., Rev. gén. des Sciences pure et appl. 36, S9 (1925).

tube. Through the bottom of this cup is sealed a tungsten wire which is he end of a small tube. An outside electr mercury is then made through this small tube by a with the tungsten wire. The bottom is closed with a tight fitting rubber hrough which pass the inlet and outlet tubes for the liquid ammonia. stopper amough which pass the line t and outlet tables for
The outlet tube extends almost to the top of the inner tu ence the cube chosen and annote to the top or the limit that the limit the method of the stop per leak p_{top} is always surfounded by the India. The proof and more rigid, it is covered with sealing wax.

The liquid ammonia is pumped from the Dewar bottle D to the antick again by a small iron gear pump (E) which is driven by $\frac{1}{2}$ is the set of $\frac{1}{2}$ and $\frac{1}{2}$ in the set of $\frac{1}{2}$ and $\frac{1}{2}$ is the set of $\frac{1}{2}$ in the set of $\frac{1}{2}$ is the set of $\frac{1}{2}$ in the set of $\frac{1}{2}$ is the set of $\frac{1}{2}$ is the set of e bottle. The temperature of the ammonia may be lowered below the boiling point by pumping off this vapor

Fig. 1. X-ray tube with a mercury anticathode.

lectron bombardment of the mercury. A nine cubic centimeter sample of the pure mercury was introduced into the mercury cup of the anticathode h the side arm F , which oil pump. The pressure was measured by a MacLeod gage. Mercury vapo by a set of two mercury vapor condensation pumps backed by a vacuum was condensed in a liquid air trap and thus prevented from entering the tube while it was in operation. The tube was excited by an x-ray transform measured by a sphere gap connected in para e closed magnetic-circuit type and the potential across the secondary wa

112,000 volts and about 1.5 milliamperes for a total of 37 hours. In a second Two experiments were performed. In the first the tube was operated at experiment it was operated at 138,000 volts and about 2 milliamperes for 81 hours and 146,000 volts and 2 milliamperes for 4 hours. The mercur rformee
nilliam
at 138,
and 2 1 ed after each experiment and no trace of gold could be found

Discussion. From the number of coulombs of electricity passed through the tube, the first experiment should have yielded 0.4 gram of gold and the second 1.42 grams if every electron had lodged in the nucleus of a mercury atom. Since the sensitiveness of the analytical method was one billionth of a gram of gold, it is evident that less than one electron in a billion was effective. Such a result may be explained in several ways. It is possible that the percentage of electrons that lodge in the nucleus is many times smaller than one in a billion and, therefore, the experiments were not conducted for long enough time. Also, as discussed in the introduction, even if an electron added itself to the nucleus the resulting atom may be unstable and, therefore, could not be detected by chemical methods. However, it seems that the failure of these experiments may possibly be due to too low a potential togive the electron sufhcient speed to drive it sufficiently close to the nucleus.

Test for Gold. In the experiments here described, particular care was taken to be sure that the original mercury did not contain gold. The mercury used was a 17 cc sample produced in this laboratory by Harkins and Mortimer' in their work on the separation of the isotopes of mercury. It had been slowly vaporized in a high vacuum and the vapors diffused through filter paper at least nine times. Previous to this treatment it had been washed in the usual way with a dilute solution of nitric acid and mercurous nitrate and distilled three times at low pressure. Only the middle portion of the distillate was retained each time. No gold was found when this sample was tested, which indicates that if any were present its amount was probably less than four parts in a trillion parts of mercury.

The method of analysis was a modification of a micro-fire assay method employed by Haber' in the quantitative estimation of gold from the water of the Rhine River. It consisted, essentially, in the collection of the gold in lead, the cupellation of the lead, and the measurement of the gold bead under a high power microscope. It was carried out as follows. The sample of mercury was distilled slowly in an apparatus of convenient size at a pressure of 0.01 mm and a temperature of 90'C. The rate of distillation was one cc in 43 minutes. When about three grams remained in the distilling flask the distillation was stopped and this residue transferred to an alundum filter crucible and dissolved in dilute nitric acid which contained neither halogens nor noble metals. After the crucible was washed with water until free from mercurous salts, it was dried and 0.5 gram each of pure lead and lead oxide together with 0.2 gram of boric acid were added and the scorification started over a small gas flame. However, the process should not be carried to completion in the alundum crucible because of the difficulty encountered later when the small lead bead has to be removed from the bottom of the crucible in preparation for the cupellation. Therefore, after the molten lead had been allowed to come in contact thoroughly with the inside of the crucible, the flame was removed and the lead button transferred to a porcelain crucible. One-half gram of lead oxide and 0.2 gram of boric

³ Unpublished work

944

⁴ Haber and Jaenicke, Zeits. anorg. Chem. 146, 156 (1925).

acid were again added and the scorification continued until the b'utton was about a millimeter in diameter. It was then removed and cupelled in a shallow, unglazed porcelain dish with a thin bottom. This dish was so thin that its contents could be examined under a microscope with transmitted light. Since the porcelain did not act as an absorbent, the gold bead remained behind in an envelope of slag. The diameter of the bead was measured with a microscope by the use of a filar micrometer eyepiece.

The porcelain dish used as a cupel was made from a small crucible cover. The handle of the cover was removed and the top ground on a glass plate with emery until it was about one millimeter thick. The glazed surface was removed by the use of a few drops of concentrated hydrofluoric acid.

The reagents, lead, lead oxide and boric acid used in the analysis were especially prepared and purified in order to be certain that they contained no gold. The lead was prepared from Kahlbaum's lead acetate.

The solution of the acetate was freed from any trace of noble metals by warming with thio-acetic acid until 10 grams of the lead from such lead acetate gave no visible sign of a residue on cupellation. The crystallized acetate was then ignited to lead in a covered crucible over a blast lamp. A portion of this lead was dissolved in pure nitric acid and the resulting lead nitrate crystallized twice. It was then heated on an electric hot plate until it decomposed into lead oxide. The boric acid was purified by twice crystallizing Kahlbaum's purest boric acid from hot water. Tests for gold were made on these reagents using lots several times greater than were employed in testing the mercury which had been bombarded by electrons.

UNIVERSITY OF CHICAGO, February 8, 1928.