

## TRANSMUTATION OF ELEMENTS

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## ABSTRACT

**Test for the transmutation in the tungsten target of an x-ray tube.**—X-ray spectrograms of the tungsten target of a deep-therapy x-ray tube were taken before and after operating it for about 80 hours at 2–3 ma and 207 kv peak voltage. No lines other than those due to tungsten were found before or after.

**Test for transmutation of lead in a lead arc.**—The experiments of Smits and Karssen with the lead arc were duplicated as nearly as possible. Under no conditions of current density was there any spectroscopic evidence of a transmutation of the lead to mercury.

**Test for transmutation of lead in a high potential discharge between lead electrodes in CS<sub>2</sub>.**—The experiments of Smits and Karssen were carefully repeated. Some evidence of Hg in the residue from the electrodes was found. The indications are however, that the mercury comes from the electrodes, the carbon bisulphide or dust particles rather than from a transmutation of lead.

## INTRODUCTION

IN RECENT years, a number of experimenters have tried to settle the important question as to whether it is possible to transmute one element into a new one by other means than  $\alpha$ -particles. It has been maintained that light elements could be transformed by means of high potential discharges<sup>1</sup> and that heavy elements could be transformed in a low potential electric arc, or when subjected to high potential discharges.<sup>2</sup> Miethe has claimed that he was able to transmute mercury into gold, and encouraged by his results, Smits and Karssen started a series of experiments by which they thought they could prove the transmutation of lead into mercury and thallium. Miethe's work has been refuted by a number of scientists<sup>3</sup> while Smits and Karssens work has not been checked.

## EXPERIMENTS WITH THE X-RAY TUBE

If it is possible to bring about a transmutation by moderate quantities of energy, one might expect to find traces of it in an x-ray tube, where high velocity electrons impinge upon a target. If any appreciable amounts of matter were transformed, it would show up in the characteristic x-ray spectrum. The evidence one might get here would be indisputable, and as

<sup>1</sup> J. N. Collie and H. S. Patterson, *Chem. Soc. Trans.*, **103**, 419 (1913); J. N. Collie H. S. Patterson and I. Masson, *Proc. Roy. Soc.*, **A91**, 30 (1915); R. Riding and S. C. C. Baly, *Proc. Roy. Soc.*, **A109**, 186 (1925).

<sup>2</sup> A. Miethe, *Naturwiss.*, **12**, 597 (1924); A. Miethe and H. Stammreich, *Zeits. f. anorg. Chem.*, **150**, 350 (1926); **156**, 185 (1926); H. Nagaoka, *Journ. de phys.*, **6**, 209 (1925).

<sup>3</sup> E. H. Riesenfeld and W. Haase, *Ber. d. D. Chem. Ges.*, **58**, 2828 (1925); E. Tiede, A. Schleede and F. Goldschmidt, *Naturwiss.*, **13**, 745 (1925); E. Duhme and E. Lotz, *Ber. d. D. Chem. Ges.*, **59**, 1649 (1926); F. Haber, J. Jaenicke and A. Matthias, *Zeits. f. anorg. Chem.*, **153**, (1926); M. W. Garrett, *Proc. Roy. Soc.*, **A112**, 391 (1926).

the tube remains closed all the time, the danger of accidental contamination is rather small.

Following this idea, which was suggested by Dr. R. A. Millikan, the author arranged an experiment in the following way: An unused, large, deep-therapy x-ray tube from Victor X-Ray Corporation was put at our disposal through the kindness of Dr. A. Soiland of Los Angeles. To make sure that no other lines except those of tungsten were present to start with, the characteristic *K*-spectrum was taken by photography, using the Bragg method. The distance between crystal and slit was the same as between the crystal and film. The crystal used was a calcite cleavage plate which was turned very slowly by means of a clockwork, a very important precaution when one does not have a very good crystal. If this is omitted, the background of the lines does not become uniform, and pseudo-lines will appear. The current was taken from a transformer and rectified by one kenetron tube. An autotransformer and resistance were used for regulation.

An exposure with 25 cm distance between crystal and film, using 83 kv and 2 ma gave a fairly good film; a new exposure under the same conditions, but with 83-85 kv gave a better film. No other lines than those of tungsten could be detected. The last exposure was taken October 16, 1925, and the tube then returned to Dr. Soiland and run by him for about 80 hours at 2-3 ma and 207 kv peak voltage. A new picture was then taken on February 3, 1926 under the same conditions as before. No lines that indicated a transmutation could be found. Later, the author had an opportunity to see a number of spectral photographs, taken with a Seeman-spectrograph of an x-ray tube that had been running for several hundred hours at around 200 kv. No foreign lines could, however, be detected.

#### EXPERIMENTS WITH THE LEAD ARC

As the next experiment was chosen that of Smits and Karssen. They have tried out different constructions of electric arc lamps, in which lead vapor acted as the conductor, and got strong evidence of a transmutation. The results obtained are in a class by themselves for the following reasons: The transmutation products, mercury and thallium were found by photographing the spectrum of the arc, keeping the lead in the same tube in which the experiment had been performed, without touching it. The other remarkable thing is that the mercury is so abundant, and yet Smits and Karssen think that if the lamp had a long enough life, a stage would be reached, when the lead spectrum would have disappeared entirely. It is evident that these conditions made this experiment a very favorable one to repeat.

The first tube constructed by Smits was shaped like an inverted *Y*. Through the two legs were sealed the electrodes, and the arc was maintained across the bend. Such a tube was also constructed by the author but failed to work satisfactorily. In the meantime, Smits and Karssen had changed the construction of the lamp, and the tube which the author built and ran successfully, was as nearly as possible a copy of the tube finally used by them. The size, however, was probably smaller because of the dif-

faculty of judging the size from their drawings and photographs. The justification for copying their tube as closely as possible is found in their statement, that small changes in the experimental conditions, may almost or entirely prevent the transmutation from taking place.

The tube used by the author was made of transparent quartz and had the following shape: (see Fig. 1.).

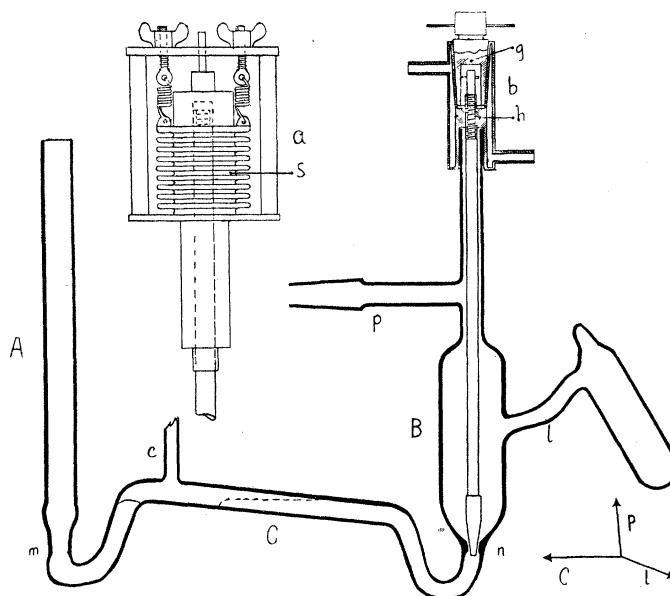


Fig. 1. Diagram of arc tube.

It consisted of two vertical legs *A* and *B*, and the connection *C*, in which the arc burned. In the two legs are inserted the rod-shaped electrodes fastened to the holder *a* (drawn separately) and *b*, which close the tube on top. The side tube *l* on *B* carries a container from which the lead used in the experiment can be poured over into the main tube. Another side tube *p* connects the lamp to the oil pump by means of a glass tube (not drawn), which is attached to the quartz tube by a cooled ground joint sealed with wax. On this glass tube are two stopcocks, the side tubes *p* and *l* are bent so that they lie in the plane of the paper; in reality they protrude as in the end view sketched on the side, where the directions of the tubes are indicated by arrows. *C* has a vertical side tube *c* which is closed by a window at the top.

The electrodes were first made of invar which was later exchanged for steel, as invar dissolves in the lead. The lower ends carry carbon points, which are turned in the shape of cones to fit the ground constrictions *m* and *n* in the quartz tube. To lift or lower the electrode in *B*, an arrangement with a cone that turns the threaded top of the electrode was constructed. The electrode holder itself is water-cooled and attached to the

tube by means of black sealing wax. This arrangement was originally used for both holders, but the strong heating of the electrode in *A* brought about some evaporation of grease from the cone. This holder was then somewhat modified, as shown separately in Fig. 1.

In Fig. 2 can be seen the tube mounted on a rectangular frame which is fastened to a heavy tripod by means of a bearing *a* and an arm *l*. This arm is excentrically connected to a pulley *b*, the axle of which is rigidly

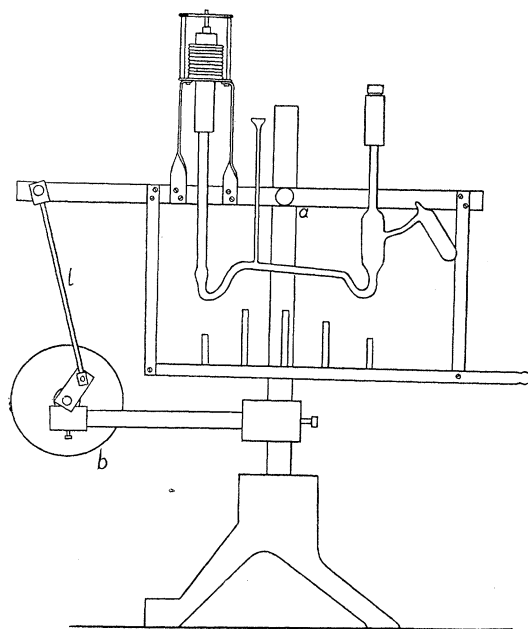


Fig. 2. Mounting of arc.

attached to the tripod. When the pulley is turned the frame is set into a rocking motion. The tube is fastened to the upper side of the frame and arranged so that it can be heated by means of 5 bunsen burners screwed on to a main tube which forms the lower side of the frame. To the frame are also fastened two copper tubes (not drawn), each having a row of fine jets directed against the quartz tube, through which compressed air could be blown to cool the tube.

For the photographing of the spectrum, a Hilger quartz spectrograph model "E 3" was used, where the spectrum from 2100 to 8000A is recorded on a 10"×4" plate. The light is either taken out directly and by means of a quartz lens focussed on the slit, or the light coming through the window on the vertical tube *c* is reflected by means of a stainless steel mirror, and then brought to focus by the lens. Panchromatic plates from Wratten and Wainwright were used.

Great difficulties were experienced in preparing pure lead. Several methods were tried out, of which the best was found to be to recrystallize

lead nitrate, c.p., twice, heat it in a quartz tube to form lead oxide, and reduce this in the same tube by means of hydrogen. Still, this did not give lead that was free from mercury. Finally the lead was sent for, which the firm C. A. Kahlbaum in Germany had prepared for Smits and Karssén, who had found it to be almost free from mercury. The spectrum of this lead was photographed, but gave strong lines of mercury and thallium, stronger than in any of the home-made preparations. As a matter of fact, this spectrogram was used as a reference spectrogram when looking for thallium and mercury lines. The preparation of home-made lead was then resumed, and by distilling off 10–30 percent in high vacuum at 1000–1100°C the remaining lead was found to be spectroscopically free from mercury and thallium.

After the tube had been carefully cleaned, it was mounted, and the lead brought into the side tube where it was kept liquid during the whole run by means of a little electric furnace. The side tube was then sealed off, and the pumping started while the charcoal tube was heated. The further stopcock was then closed, and the charcoal cooled by liquid air. Then the frame was tipped so as to let the lead run into the main tube. The volume was regulated so that the lead filled up the tube as shown by the dotted lines in Fig. 1. A big direct current generator furnished the power, and after fastening the electric leads, so that  $b$  is the positive pole, the arc was started.

As has been mentioned before, the construction and the mounting of the tube are in as close agreement with the work of Smits and Karssen as could be judged at that time (fall, 1926). According to their statement, a low current density has no influence upon the spectrum, while a high current density may bring about a transmutation. They considered that they proved this by taking photographs of the spectrum before and after a run of 10 hours, at about 10 amp. No change in the lead spectrum could be observed. But by running the tube at 30–35 amp. and about 80 volts for 6 hours, the mercury lines begin to appear, and after 10 hours burning, the strongest mercury lines could be seen very clearly, and also the characteristic thallium lines. This and other experiments showed, that high current density was favorable for bringing about a transmutation, and they tried currents up to 60 amp. This was, however, rather hard on the quartz tubes. Consequently they changed their working conditions, and the construction described above is very similar to theirs. The idea is that the arc does not burn continuously, but flashes of high current density are formed when the lead surfaces are brought together by rocking the tube. At make of the arc, the current in the tube was 60–100 amp. and at the break considerably more. When this procedure is used, it is necessary to take the light out through the vertical tube, as a black film appears inside the tube that cuts down the intensity of the light. With this procedure, Smits and Karssen got strong evidence of a transmutation. After 9–1/4 hours of sparking all the mercury lines, even the very weak ones were present on the plate.

The author first made a run for 10 hours with continuous arc and low current density. The arc burns very nicely with a length of 5–7 cm at

12–14 amp. and 23–26 volts, and with some heating by the gas burners. No change in the spectrum was observed, in accordance with Smits and Karszen.

Then comes the question of running the tube with higher current densities. This means that the gas burners must be put out, and cooling applied, so as to get rid of the heat developed in the arc. But the cooling must not be too strong, because the lead must be kept liquid all the time. If it solidifies, the tube will usually break when cooling down, and invariably during reheating.

After several trials, a run of 35-1/4 hours was completed. At the make of the arc the current was 60–75 amp. for 14 hours, and 80–90 for 21-1/4 hours. Of this time about 20 percent was effective burning time for the short flashes. The amount of lead used in this experiment is only 180–200 grams, and it was found to be spectroscopically pure before the run was started. The spectra throughout the experiment were mostly photographed with 5 hour intervals under standard conditions with 12–14 amp. and 25 volts in vacuum, and showed no mercury or thallium lines. On trying to run the tube with continuous current, it broke down, because of too strong cooling.

The tube was then repaired, and a new run started with new lead, which, however, on very good spectrograms showed faint traces of mercury and thallium. Since Smits and Karszen had reported that they also had had success when the tube was filled with nitrogen, such an experiment was tried. The tube was first thoroughly evacuated and pure nitrogen let in, so that the pressure was 1/5–1/7 of an atmosphere. The length of the arc was regulated by means of cooling and heating the charcoal tube, which then took up or gave off nitrogen, and by this change in pressure, the level of the lead could be controlled.

First, an experiment with continuous current was made for 25 hours, with current densities from 15–25 amp., the latter value being maintained for 10 hours at about 38 volts. No appreciable change in the intensities of the mercury and thallium lines could be observed.

With the same tube, the method of flashing was again taken up. Accidentally it had been found, that an oscillatory motion of the lead in the short leg of the tube can be established at a certain pressure. If the two inner lead surfaces are joined, an arc is formed, which heats up and expands the nitrogen, so that the distance between the two surfaces increases very rapidly, pulling the arc out to a length of 12 cm, where it breaks. Now the nitrogen between the two lead surfaces cools off, which brings about a new contact, and a new arc is formed. These conditions were made use of in the following run, and did away with the rocking device. The pressure was again regulated by means of the charcoal tube. The actual burning time of the intermittent arc was a little less than half of the total time, and each flash lasted about half a second. After a run of 10 hours duration with a current of from 65–95 amp. at make the scheme of connections was changed, a condenser was placed across the terminals and a big inductance

in series with the tube to protect the generator against transients. With this arrangement a 12 hour run was taken the current being raised to 120 amp. in the make, but no increase in the strength of the mercury and thallium lines could be detected. On increasing the current to 150 amp., the tube broke after 2.5 hours run, but there is no doubt, that a new tube would stand that current for 5-10 hours.

As will be seen, the currents compare with and even exceed those used by Smits and Karssen, and as the diameter of the tube where the arc is burning is only  $3/8$ " , the actual current densities are higher. On account of the smaller amount of lead used (180-200 g against  $\pm 900$  g), the expected products of transmutation should be more easily detected.

#### EXPERIMENTS WITH DISCHARGES IN CARBON BISULPHIDE

It has been mentioned before that Smits and Karssen have found that small changes in the experimental arrangements may destroy the transmutation. They have, however, also reported a second method of producing a transmutation, and they state that by this method it is possible to get *reproducible* results. The process is as follows<sup>4</sup> (see Fig. 3.): Two lead elec-

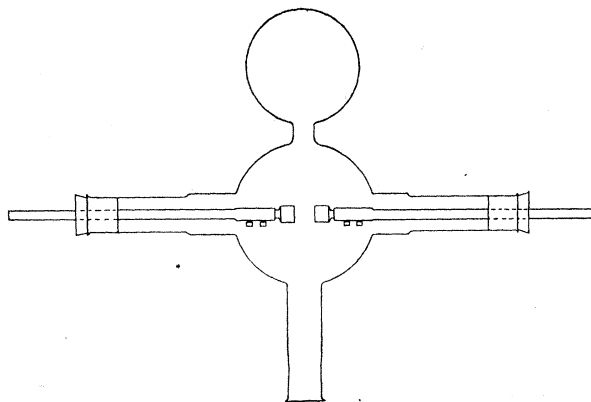


Fig. 3. Tube used for high tension discharges in  $CS_2$ .

trodes in steel holders are placed horizontally and opposite each other in a Pyrex flask. The holders are stuck through cork stoppers and are, on the other side, connected to the electric leads. The neck is closed by a stopper, through which is inserted a long glass tube, which acts as an air cooler. The flask itself is filled with carbon bisulphide. High potential discharges are passed between the lead electrodes, the surface layers of which are torn off and go out in the solution as a fine suspension which precipitates and collects in the bottom in addition to carbon and sulphur which are formed by the disintegration of carbon bisulphide. The precipitated material is collected and analyzed chemically, and mercury is found. As will be seen, this method is much less clean-cut than the previous one, as the system is

<sup>4</sup> Smits and Karssen, *Zeits. f. Elec. Chem.*, **32**, 577 (1926).

not quite closed, and the mercury is found by a rather complicated chemical analysis. Another objection is that it would be very difficult to perform a reliable chemical analysis of the carbon bisulphide if one wanted to make sure that even small traces of mercury are absent. Nevertheless, the experiment was tried.

A tube like the one drawn was made, but instead of steel holders, glass tubes filled with lead were used in the first experiment. Instead of the smaller bulb underneath the main tube, a cylindrical container was used in the first experiment, similar to a test tube, in accordance with Smits and Karssen's design. Here the sputtered lead is precipitated and accumulated. The current used was taken from a set of three transformers from the Roentgen Manufacturing Co., San Francisco, one insulating transformer and two "boosters" furnished with a mechanical rectifier. Over the tube was put a screen cage for protection in case of fire.

Carbon bisulphide, quality c.p. from Baker, was used in the first experiment, after it had been dried over calcium chloride and re-distilled. In the later experiments Anchor Brand, commercially, guaranteed not less than 99.9 percent  $\text{CS}_2$  from Wheeler, Reynolds & Stauffer, San Francisco, was used. This product was distilled once with a high rectifying column, dried over chloride and again distilled in the same apparatus. The reason for using this commercial product is a statement in the literature that mercury is sometimes used in the preparation of c.p. carbon bisulphide, which then might be contaminated with mercury. In both distillations only 4/5 of the whole volume was driven over, since there was a possibility that the heavier end-fraction might contain mercury compounds. The five percent distilling over first was also thrown away.

The runs were usually of 5–12 minutes long, separated by 20–40 minute intervals to let the carbon bisulphide cool down. In this and the following experiments the current was from 20–40 ma. The potential difference also varied considerably, but averaged 148 kv peak, measured with sphere gaps.

The first run lasted, when the intervals were added up, for 1 hour 20 minutes. The precipitated material was brought out of the tube and dried. It amounted to 7 gr, but held a fair amount of sulphur. This together with 3.5 gr, cut loose from the rough surface of the electrodes, was subjected to the same kind of chemical analysis as employed by Smits and Karssen. The method is mainly built upon a procedure described by Stock and Heller.<sup>5</sup> To separate the mercury from the bulk of the lead, the precipitation is brought into a quartz tube, which is heated to 800–900°C with a slow stream of hydrogen going through. The hydrogen takes the mercury along with it to a double U-tube where the latter is condensed, and by means of chemical methods one finally gets out the mercury in metallic form. It can also be converted into mercury iodide which forms easily distinguishable red crystals. An amount of 1/100 of a milligram of mercury can not be overlooked, and amounts of 1/1000 of a milligram of mercury as iodide can be rather easily detected under the microscope.

<sup>5</sup> Stock and Heller, *Zeits. f. angew. Chem.*, **39**, 466 (1926).



The result of the first run was an amount of mercury between 1/10 and 1/100 of a milligram. To make sure that there was no mercury in the electrodes, they were analyzed by melting them down and heating the lead in a stream of hydrogen, proceeding in the same way as with an analysis, but no mercury was found. Blind tests showed that no mercury came in through the chemicals used, and numerous tests with solutions of 1/10 to 1/100 mgr. of mercury, treated as in the analysis, showed, as far as could be judged, that the amounts could be recovered.

In the next experiment the above mentioned carbon bisulphide was used. To ascertain the purity of the lead, hydrogen was bubbled through it for 24 hours at 850°C and for 12 hours at 950°C. The purity was tested by heating it in the same vessel in a stream of hydrogen, and analyzing the distillate. No mercury could be detected. Then the actual run was started and a total time of 2 hours was attained. The amount of precipitated material amounted to 4.5 gr which, together with 3.5 gr from the electrodes was distilled and analyzed as before. The result was a very evident positive mercury reaction, but the amount was much less than before.

At the same time, a purification of the German lead from Kahlbaum was carried out by heating it for 20 hours at 950°C while hydrogen was bubbled through. Then the previously mentioned double U-tube was melted on, and an analysis performed, the difference from the previous analysis of lead for electrodes being that the hydrogen actually bubbled through the molten lead. This analysis gave mercury, although not very much. This seemed to show, that the procedure of leading hydrogen over the lead was an unreliable process of analysis, and the bubbling of hydrogen through the molten lead could not be relied upon for purification, when amounts of a hundred grams are involved. The electrodes used in the last experiment were then melted down, and an analysis performed by bubbling hydrogen through the liquid lead. This analysis, however, gave no mercury.

Previous experiments had shown that it was possible to get rid of the mercury by distilling it off in a high vacuum. Consequently half of the German lead in the container was driven over, and the residue was analyzed by leading hydrogen close over the surface of the lead. Due to experimental difficulties it was not possible to let the hydrogen bubble through the lead this time. This analysis gave no mercury. With this lead a run of 3.5 hours was performed, yielding about 11 gr of washed precipitate. This was analyzed in the usual way and gave only a slight mercury reaction.

The results of these runs seem to indicate that with a sufficient purification of the materials used, it is possible to reduce the amounts of mercury found, which again means that the mercury originates from electrodes, carbon bisulphide and dust coming in during the experiment. In a physical laboratory it is very hard to find places which are free from mercury. All dust must be suspected of containing it, and this necessitates an extreme cleanliness in handling all objects used.

## CONCLUSIONS

As will be seen, the results of the experiments with the x-ray tube and also with the lead arc lamp are absolutely negative, although the extremely small possibility exists that Smits and Karssen chanced to establish some extraordinary experimental conditions whereby a transmutation is possible.

The experiments with the discharges in carbon bisulphide are not quite negative, but it is the authors opinion that it would be possible to get clean negative results with quite pure substances and under conditions where no accidental contamination was possible.

The author wants to express his gratitude to Dr. R. A. Millikan for helpful suggestions during the work.

In a letter from Smits<sup>6</sup> to "Nature" which appeared after this work had been finished, he states that the greater amount of mercury which he got in the experiments with discharges in carbon bisulphide, is due to contaminations. But he maintains that there is still a residual effect which can not be ascribed to that cause. In a later article by Smits and Frederikse<sup>7</sup> they describe a repetition of the experiment of Smits and Karssen with high potential discharges in carbon disulphide, in which their results were entirely negative. They think that the positive results previously obtained must be ascribed to impurities in the carbon disulphide used in the first experiments.

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<sup>6</sup> Smits, *Nature*, **120**, 475 (1927).

<sup>7</sup> A. Smits and W. A. Frederikse, *Zeits. f. Elektrochemie* **34**, 350 (1928).