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A MASS SPECTROMETER FOR ROUTINE SOLID ANALYSES

by

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I. INTRODUCTION

Originally the plans for building a mass spectrometer for routine analyses at the Idaho Chemical Processing Plant, then operated by the American Cyanamid Company, were based on an instrument similar to those used in Argonne National Laboratory, basically identified as the MA-15 model.¹ It was requested that the instrument incorporate a vacuum lock similar to that developed by this Laboratory.² At that time there were available complete drawings for such an instrument and, insofar as material would permit, fabrication in the ANL shops could have been started immediately.

However, it was felt that numerous improvements would be desirable. As a result, a practically new instrument was evolved. A complete new set of drawings had to be made.

In designing this instrument the objectives were to:

1. Incorporate the latest features and improvements,
2. Engineer it for safe and reliable operation,
3. Make it reasonably attractive in appearance.

The purpose of this report is not to give a complete technical description of the instrument but mainly to describe the principal features and the results of its operation before the instrument was turned over to the Idaho Chemical Processing Plant.

II. INSTRUMENT

The basic instrument is a 12 in., 60° type and is similar to that developed by Inghram. The tube, magnet, and vacuum system are mounted in a cabinet with the electronics located in relay racks as shown in the overall view in Figure 1.

¹To be published.

²C. M. Stevens, Rev. Sci. Instr. 24 148 (1953)

From the beginning it was planned that an improved design of the vacuum lock, based on the experience with the first version, was to be made. The mistake of having similar metal surfaces in sliding contact was to be avoided. The final choice was the use for the manifold of a non-magnetic 300 series stainless steel, hardened by the Malcomizing process, and, for the carriages, of chromium-plated 300 series stainless steel. One of the carriages is shown in Figure 2.

The lengths of the carriages and channel are considerably shorter than in the first model. This is a result of cupping the blank carriage at both ends to enclose the source assembly on the end of the sample carriage, thus making possible shorter sections between stages. After the sample carriage is inserted, the cupped blank carriage is pulled out enough to expose the source assembly inside the source head.

The vacuum lock on this instrument has a means for supporting the carriage which is pushed out when the other carriage is inserted from the opposite end. This consists of a hook mounted on each end of a 1 in. shaft above the vacuum lock to which the carriage is attached before inserting in the vacuum lock. In Figure 1 the carriage on the left is attached to the hook and is ready to be inserted. Figure 3 shows the carriage inserted and ready for an analysis.

The analyzer tube is 2 in. O.D., 1/16-in. wall Inconel as compared to 1-1/2 in. O.D., 1/8 in.-wall copper in all previous 12-in. radius tubes. This diameter allows a wide mass range in using the ratio recorder and gives high pumping speed to the source region, which is the origin of gas when using the vacuum lock. This larger diameter tube required the construction of another jig for flattening the tube. This jig was the major cost in the fabrication of the tube itself. The tube is all heli-arc welded, on the inside where possible, and normalized. The collector assembly, Figures 4 and 5, Kovar lead-in seals, and slit adjuster are all mounted in a removable flanged head, and only a plate is welded to the tube. Thus, the unit can be easily removed from the instrument for any repair work. Also, a longer head can be made to mount an electron multiplier collector assembly and be interchanged with the existing head. An improved mounting of the collector end of the tube is used with a pivoted bracket, allowing motion only in the direction of thermal expansion of the tube; in all other directions the mounting is very rigid.

The source head, including the flanged plates for mounting the vacuum lock, is flanged to the analyzer tube. This permits interchanging any type of source head easily without any unsoldering or removing the tube. The two ground potential defining slits are each 0.006 in. and are mounted on the end of the 2-in. tube as in standard practice.

The surface ionization source is identical to that used in the ANL vacuum lock instrument and is based on previous designs.^{3,4}

The magnet is a permanent magnet type designed by Inghram and Stanton. The field between the 7/8-in. pole gap is varied by a shunt whose distance from the pole faces is changed by a worm and gear drive, Figure 3. The Alnico-V, after assembly in the yoke, is magnetized by a set of 30 coils each having 43 turns of 3/8 in. x 0.050 in. double Formax cotton-covered copper strap. During the magnetizing, the shunt is used to short out the pole gap reluctance. With a total magnetizing force of 100,000 ampere turns, a field in excess of 9,000 gauss was obtained in the pole gap. However, after the shunt had been driven back and forth several times, this dropped to 8,000 gauss. With a 12-in. radius tube this field will bring in ions of mass 270 (UO_2^+) at 12,000 volts. With the shunt separated from the pole faces by 1/16-in. brass spacers, the minimum field is about 1300 gauss. With the spacers removed and the shunt in contact with the pole faces, the magnetic field is about one-tenth the maximum field, or 800 gauss in this case. These measurements were made on another, identical magnet.

For scanning of the magnetic field, a set of 10 auxiliary coils of 750 turns of #24 gage copper wire is used. At 250 ma these provide an exterior magnetizing force of 1,875 A.T. and a change of 1% in the magnetic field or 2.0% in mass. The range corresponds to about 6 mass units at mass 270 (UO_2^+). The sweep circuit, supplying this set of shim coils in series, will change the current from 0 - 250 ma over a period of 0.088 to 32 min.

The analyzer, source, and third stage of the vacuum are each pumped by a D.P.I. Model GHG-15 mercury diffusion pump with a trap having a speed of about 20 liters. These systems are connected to the corresponding pumping leads using a 2-in. glass ball and Inconel socket waxed together with Apiezon W wax. The metal sockets were lapped to fit each glass ball. The analyzer and source diffusion pumps are backed by one Welch Type 1405 mechanical pump. Another Type 1405 backs the third diffusion pump on the third stage. The first and second stage of the vacuum lock are each pumped by a Welch Type 1402 mechanical pump. Type VG-1A ionization gages are used to measure the pressures in the analyzer and source systems, while a Phillips gage is used on the third stage. A single elevator assembly is used for raising and lowering the three 8-liter thermos flasks around the traps. The elevator is operated by a hand crank driving a rack and pinion with a cable and pulley system.

³A. J. Dempster, Phys. Rev. 11 316 (1918)

⁴M. G. Inghram, Advances in Electronics, edited by L. Martin (Academic Press, 1948), Vol. 1, pp. 219-268.

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The frame and cabinet for the instrument was custom made from a design by C. Bullinger of the Central Shop Engineering Design Group. It is made in two sections for easy removal of the magnet. The cabinet is entirely enclosed with removable panels and grilled doors. Diagonal braces (not shown in the photographs) were put in the front and back of the frame to increase the rigidity.

The ratio recorder system was based on a similar unit that had been installed on one of our instruments. Since then this system has been incorporated in one of the ANL instruments.⁵ Essentially, it consists of a grid collector placed in front of the usual collector or exit slit. The grid was made of 0.002-in. diameter tungsten wire spaced 0.006 in. apart according to the technique described by Glenn.⁶ In addition to the usual secondary electron suppressors placed in front of both the grid collector and single slit collector, another suppressor is placed between the grid collector and slit plate, and the slit plate is insulated so that a voltage may be applied to it. Voltages are supplied by batteries to the three suppressors and slit plate to eliminate extraneous effects due to secondary electrons and positive ions. The signal to the grid collector is measured by a high gain negative feedback D.C. amplifier with a 4×10^{10} ohm grid resistor and recorded by one pen in a 2-pen Brown Electronik Strip Chart Recorder. This grid collector has an effective slit width of $3/4$ in., which will collect about a 6% range of mass at any time. Thus, a 3% range may be scanned by the single slit collector and maintained on the grid collector at all times. The signal to the single slit collector is measured by an Applied Physics vibrating reed electrometer using a 2×10 ohm grid resistor and recorded on the other pen of the 2-pen recorder. The grid collector signal can be substituted for the battery voltage supplying the potentiometer of the second pen, thus making the recording of the ions collected at the single slit collector proportional to the ions intercepted by the grid and not dependent on intensity variations.

All the electronic units, with the exception of the Applied Physics Vibrating Reed Electrometer, were designed and built by the ANL Electronics Shop. These units were identical to those built for the ANL instruments. The magnet sweep circuit, already mentioned, incorporated a "boot-strap" sweep controlling a voltage regulator to give a linear rate of change of current. The grid collector signal is measured by a D.C. negative feedback current amplifier which will deliver 0 - 10 ma. This unit also incorporated the circuit for ratio recording.

⁵C. M. Stevens and M. G. Inghram, Rev. Sci. Instr. 24 987 (1953)

⁶W. E. Glenn, AECD-3337, January 3, 1953.

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A system of protective devices are incorporated in the water system supplying the water jackets of the diffusion pumps. This system automatically turns off the diffusion pump heaters in case of water supply failure, and in case of a break in the water line in the instrument, will shut off the water supply and diffusion pumps.

III. INSTRUMENT OPERATION

After the construction and assembly, the instrument was operated and thoroughly tested for about a month before being disassembled and shipped. The instrument performed excellently from the first, and only minor adjustments were necessary. One was in the collector assembly to perfect the new technique of ratio recording being tried for the first time on this instrument.

The analyses of uranium samples by the conventional direct recording method agreed with the accepted values well within the limitations of the method of surface ionization. Two analyses of enriched uranium and two of normal uranium demonstrated this.

In the beginning the analyses determined by ratio recording were in error. Enriched sample results were 2-3 % low in the U^{238}/U^{235} ratio and 2-4 % high in the U^{234}/U^{235} ratio. Normal uranium results were about 1-2 % high in the U^{235}/U^{238} ratio. A study of the recordings indicated these errors could be accounted for by a regular variation in the grid collector signal even with a constant intensity ion beam.

Just what caused the grid to receive a different ratio of the ions depending on the small variations in ion beam position is not known. In the belief that these errors were due to secondary electrons or ions, the effects of the suppressor voltages were studied a great deal. Also, the collector assembly was modified to insulate the slit plate in order to be able to apply a small voltage to it. It turned out that adjustment of the magnet, principally for centering the ion beam in the tube in a direction parallel to the magnetic field, eliminated the errors, at least to the presently accepted limitation of the method of surface ionization, which is $\pm 1\%$ in the ratio.

All the test analyses were made with a standard sample load of 50 μg . At the start many of the runs did not give high enough intensity, but it is believed this was because the filament was not located properly. By cut and try, the best filament position was found such that it was possible to obtain consistently high intensity runs. Several of the runs gave good intensity much longer than had been obtained in other instruments.

The resolution and peak shape was fully equal to the performance of any of the 12-in. instruments at ANL. The background one mass unit away at mass 270 was about 0.01 % at an analyzer pressure of 3×10^{-7} mm Hg.

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With source slits of 0.006 in. and a collector slit of about 0.15 in. the peaks showed a long flat top with a normal sharp rise on the sides.

The entire vacuum system performed very satisfactorily. The pressure obtained in the analyzer during the test period was 3.0×10^{-7} mm of Hg, which is more than adequate for routine analysis.

The vacuum lock unit on this instrument proved to be successful. This unit has been designed with standard bearing clearance (0.0005 in.) and tolerance (0.0001 in.) with the hope of eliminating the need for rubber "O" rings. Without the "O" rings the pressure in the first stage was less than 0.5 mm of Hg using a model 1402 Welch Duo Seal Pump (100 l./min.), and the pressure in the source showed no change on sealing the "O" rings, which indicated negligible inleakage beyond the second stage. No galling or seizing of the parts was experienced at all during an estimated 50 operations of the unit, proving the choice of metal surfaces in sliding contact.

The permanent magnet proved to be satisfactory in every respect. The resulting range of field strength was 800 - 8,000 gauss. The excellent resolution and peak shape demonstrate the satisfactory design of the over-all unit. No difficulties were encountered in the operation of the mechanism for moving the shunt. The sweep coil system fell short of expectation in the range of magnetic field and mass that could be covered, but was sufficient for the routine analysis of uranium and plutonium.

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Figure 1

Over-all View of Spectrometer and Controls

Figure 2

Vacuum Lock Sample Carriage

Figure 3

Front View of Instrument Showing Source Region and Vacuum Lock.

Figure 4

Top Region of Instrument Showing Permanent Magnet and Collector Assembly

Figure 5

Rear View of Permanent Magnet and Collector Assembly