Feb. 4, 1964 R. E. FEARON 3,120,610

APPARATUS FOR PRODUCING A HIGH INTENSITY ELECTRON STREAM USED TO EXCITE CHARACTERISTIC RADIATION OF ELEMENTS

Filed May 15, 1961 2 Sheets-Sheet 1

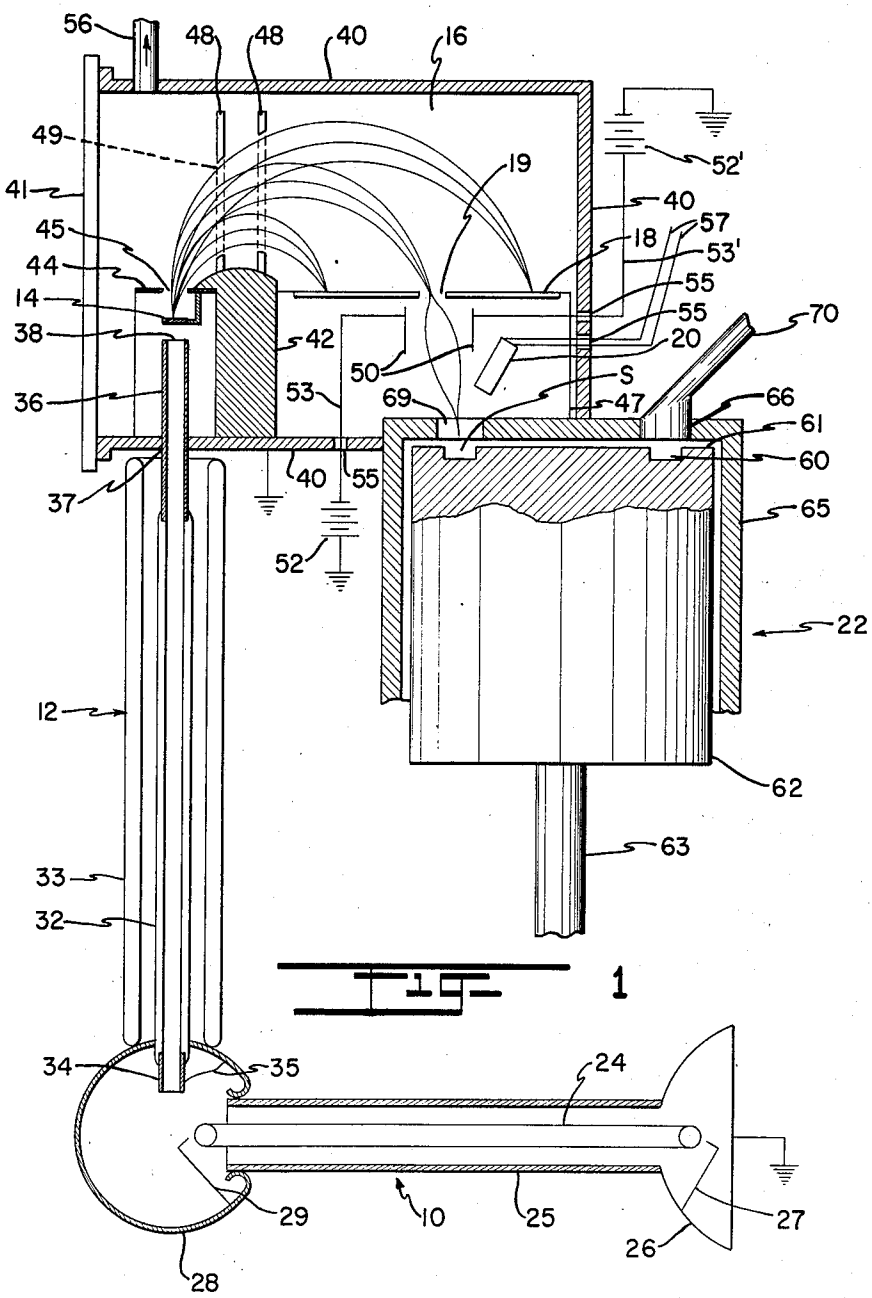

Fig. 1

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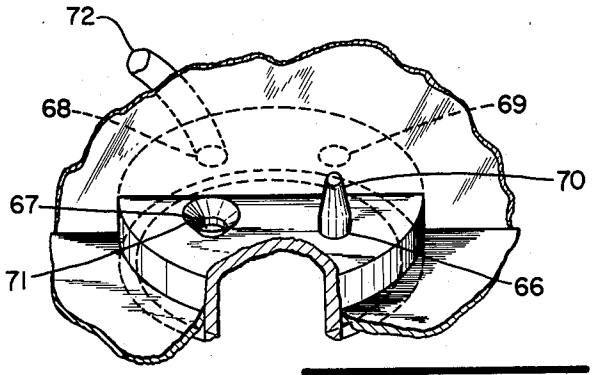

FIG 2A

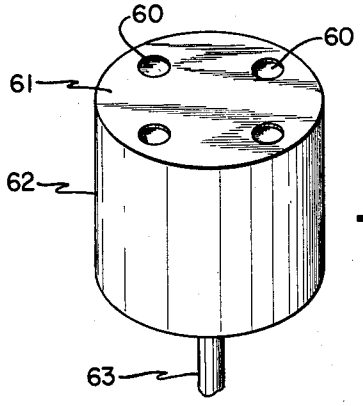

FIG 2B

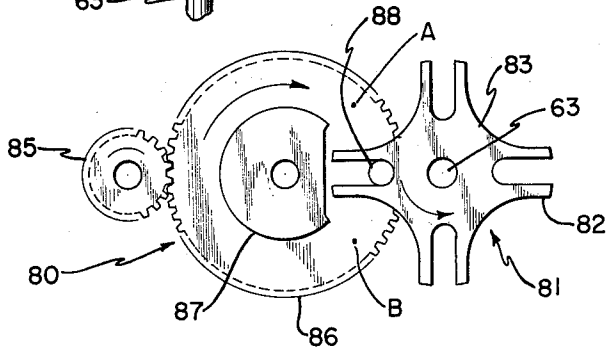

FIG 3

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United States Patent Office 3,120,610 Patented Feb. 4, 1964

3,120,610

APPARATUS FOR PRODUCING A HIGH INTENSITY ELECTRON STREAM USED TO EXCITE CHARACTERISTIC RADIATION OF ELEMENTS

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Filed May 15, 1961, Ser. No. 110,174
8 Claims. (Cl. 250—49.5)

This invention relates to a novel and improved method and apparatus for radiological analysis, and especially for quantitative analysis of elements; and, more particularly, relates to a way of producing a particle stream of high intensity electrons at a controllable energy level for impingement on a substance in order to excite characteristic radiation from known elements present in the substance, together with improved means for handling substances for analysis.

In my copending application, entitled "Apparatus and Method for Conditioning and Analyzing Earth Components," Serial No. 586,172, now Patent Number 3,031,571, issued April 24, 1962, the invention disclosed therein included as one stated objective the analysis of elements which are common low atomic number constituents of the earth by exciting their characteristic radiations associated with K shell transitions. In relation to that case, it was proposed to use streams of material particles, as distinguished from for example, electromagnetic radiation, for the reason that a larger proportion of the produced radiation from a sample is, in fact, characteristic radiation and for the further reason that the radiation producing interaction of such streams with matter is very much stronger than the interaction of corresponding electromagnetic radiation with low-density substances, such as air-drilled cuttings removed from a well bore. Thus, it becomes possible with streams of material particles to analyze smaller, low atomic number samples of matter.

In the design of some suitable means and method for exciting characteristic radiation from elements, and particularly to the end of quantitatively analyzing such elements, consideration must be given to the energy required for excitation of radiation from the various elements, keeping in mind that this energy level will vary depending upon the element to be analyzed, and in general, the highest energy of characteristic radiation would be in the range of 120 electron kilovolts. Also to be considered is the problem of energy absorption and other losses by the source of particle radiation, as well as the physical density of the material. Relative to the latter considerations, provision must be made for a high degree of directional selectivity of the electron beam in order to isolate the characteristic radiation of the particular element to be analyzed, particularly since the continuous spectrum of target radiation is emitted in the equatorial plane of the direction of propagation of an incident particle stream, whereas characteristic radiations are emitted equally in all directions. Directional selectivity is also important for the reason that the directiveness of the continuous spectrum is especially strong for low-energy electrons with respect to their target radiation. Accordingly, it is necessary to define a radioactive emitter as something approximating a "point source" in terms of the practical geometry of experimental equipment. Slits, apertures, collimating means and other means of controlling or directing electron emission from a small source must be physically larger in dimensions, so that the source appears to be a point in comparison to the dimensions of the slits, etc. In any event, it is desirable that the controlling means be close to the source and also be scaled down to small physical dimensions; this in turn leads to the requirement that the impinging electron beam

be highly concentrated and that the source-emitting spot be correspondingly small.

In addition to the above, in the analysis of elements of different atomic number, each requiring a different energy level for characteristic radiation, the electron beams must be closely, accurately controllable both in strength and direction so as to exactly correspond with the energy of characteristic radiation of each particular element while compensating for any losses, for instance, due to self-absorption or collimation. Here, an important factor is to make provision for some means of delivering an electron beam of the necessary strength and intensity coupled with some means for properly focusing it on the source; and, in this connection, it is highly desirable that the equipment employed be capable of accomplishing this in a practical way, and specifically so as to be economical and of a practical size for convenient use in the field. Also, the equipment employed must be safe and dependable in use and particularly to obviate any danger of radioactive contamination.

In proposing the use of a concentrated electron beam which may be accurately focused on a limited area, for example on a sample, distorting influences of the surrounding environment must be removed and, as discussed in detail in my copending application, the space selected for analysis must be evacuated at least to the extent that as far as the travel of the electron beam is concerned, a perfect vacuum will exist. In other words, it is necessary to minimize the probability that the electrons will undergo deviation or that their range will be restricted due to collisions with the environment. For this reason, the present invention also contemplates a way of transferring samples brought into the analyzing space in a continuous manner without affecting the vacuum established, and while avoiding the use of special entraining substances which are expensive and somewhat difficult to handle. In this connection, the existence of a vacuum is particularly important because it makes possible the use of low-energy electrons as the entire means of exciting the sample. Low-energy electrons have even less range in air than does electromagnetic radiation which corresponds with their energy. Thus, a 10 kilovolt electron will have a range of less than $\frac{1}{20}$-inch in air whereas the corresponding electromagnetic radiation may very well penetrate a few inches of air before being totally absorbed. Moreover, due to the very high absorbability of electrons, there is a direct benefit accruing from the fact that the electrons can be employed to analyze very thin layers of materials, comprising almost imperceptibly small samples. A layer of a solid .001-inch thick is more than adequate to afford a complete opportunity for sample analysis by way of the electron excitation method to be described. However, it is not possible to employ such highly absorbable radiations and derive the advantage which is a direct consequence of their high absorbability without providing a suitable optical path to handle their necessary travel through space, to and from the sample being analyzed; and such a path must be, among other things, devoid of excessive absorption due to the presence of air or any gas of appreciable density.

Accordingly, the present invention represents a substantial improvement over my earlier method and apparatus, set forth in the hereinbefore referred to copending application as well as other known methods and apparati, and essentially relates to a way of analyzing substances to determine the amount of a selected element incorporated therein by introducing the substances into a controlled, evacuated environment, bombarding the substances with electrons at the necessary energy level to excite characteristic radiation from the selected element, and then analyzing the emitted rays quantitatively. Specifically,

this invention represents an improvement over my earlier method and apparatus in that it is adapted for use in exciting characteristic radiations of virtually any selected element, including the low atomic number elements, by provision for a practical and economical way of delivering and focusing selected electron beams for impingement on the sample, determining therefrom the amounts of known elements present in the sample, and additionally by providing for an improved way of introducing and removing samples from the evacuated space for analysis.

It is therefore a principal and foremost object of the present invention to produce an abundant source of electron particles in the form of a highly concentrated, directed beam which is selectively controlled for impingement on a sample to be analyzed for exciting characteristic radiation of a selected element therein.

It is another object to make provision for the delivery and focusing of a selected and easily controlled kinetic energy in a particle stream of electrons for impingement on a sample for quantitative analysis of a selected element therein; further, to provide for the accurate continuous analysis of such elements through a unique succession of steps involving the determination of a discontinuity in the function of radiation output from the samples versus kinetic energy per particle for impinging electrons.

It is a further object to make provision for a method of accomplishing the preceding objects in which delivery of the particle radition is separately controllable, whereby the intensity of electron particles can be regulated independently of the energy per electron contained in the beam, and vice versa.

It is a further object to make provision for the delivery of electron particle beams in such a manner that other radiation consequent to the production of such particle streams is absent, together wtih a way of closely and accurately controlling the energy level and direction of the beam so as to exactly correspond with the energy of characteristic radiation of the element to be detected while compensating for self-absorption and collimation losses.

It is a still further object to make provision for means whereby electron particle streams may be produced and utilized in an evacuated space which, so far as the electron beams are concerned, acts as a perfect vacuum to avoid deviation or deflection of the beams as they are concentrated on the sample, and where a vacuum may be maintained within the space in a simple and economical manner with conventional equipment.

It is a still further object to make provision for a source of high-potential energy which is economical and of a practical size for delivery of a highly-concentrated, electron beam capable of being absorbed and scattered to produce a spectrum of electron beams within the required energy range for quantitative analysis of virtually all known elements, and moreover to make provision for means of selectively focusing a beam of the desired energy level on a sample containing the selected element for quantitative analysis thereof.

It is an additional object of the present invention to provide for a method and apparatus of introducing a sample of material to be analyzed into an evacuated space, at the same time preventing the occurrence of any large inrush of air into the evacuated space, removing the sample from the space and replacing the removed sample with a succeeding new sample, all while preventing the large inrush of air into the space and while accomplishing a number of successive operations in a rapid and continuous manner; moreover, to make provision for a method and apparatus whereby the air originally surrounding and absorbed in a sample to be analyzed may be separately removed through a low-grade vacuum system before the sample is brought into the space where a more stringent vacuum is maintained and where the analysis is actually performed, and similarly where in transferring the sample from the low-grade vacuum into the high-grade vacuum region, no opening is created which at any time would permit the introduction of residual gases or air from the low-grade vacuum region into the high-grade vacuum region.

The above and other objects, advantages and features of the present invention will become more apparent from a consideration of the following detailed description, taken together with the accompanying drawings, in which:

FIGURE 1 is a somewhat diagrammatic view of a preferred form of sample analyzing apparatus, in accordance with the present invention.

FIGURE 2*a* is a more detailed, somewhat perspective view of a fragmentary part of the sample feed system employed in the preferred form of apparatus illustrated in FIGURE 1.

FIGURE 2*b* is a view similar to FIGURE 2*a* illustrating another part of the sample feed system.

FIGURE 3 is a plan view of a suitable mechanism which may be employed in driving the sample feed system of this invention.

Referring in detail to the drawings, there is shown, for purposes of illustration, in FIGURE 1 a diagrammatic representation of a preferred form of apparatus for conducting quantitative, radioactive analysis of a selected element which, for example, is contained in a sample, indicated at S. Broadly, the apparatus of this invention makes possible the accurate quantitative analysis of a sample by producing a high potential energy source, for example on the order of one-half million to one million volts by means such as the high-voltage generating apparatus **10**, and from this source electron particle beams are accelerated through a discharge tube indicated at **12** into a highly-concentrated beam of a known energy level. This beam, developed in the manner described, is of course at a kinetic energy level much above that required or even capable of being utilized for radioactive analysis; however, by direction of the beam through some absorbing medium such as the material absorber **14**, it is possible to scatter the concentrated beam while absorbing a substantial amount of the energy so as to substantially reduce its maximum energy level. Essentially, depending upon the characteristics of the absorber, especially its thickness and density, it is possible to reduce the maximum energy level down to the maximum energy requirements for analysis of the sample S. Moreover, depending upon the energy of characteristic radiation of the element to be analyzed, electron particles of the desired kinetic energy may be focused accurately and in the form of a highly-concentrated beam on a very limited portion of the sample and at the resonant efficiency level required for characteristic radiation from the element to permit accurate measurement of the quantity present. This focusing is preferably accomplished through the utilization of a magnetic field passing perpendicular to the plane of the drawing, through analyzing space **16**, and according to the strength of the field will cause the scattered particle beams passing through the material absorber to follow a substantially semi-circular path through the magnetic field and to strike a plate **18** at different points according to the energy or velocity of the particles. By controlling the strength of the magnetic field and by arranging an aperture **19** in alignment with the sample S, particle beams of a selected energy can be directed through the aperture to focus on the sample S, the energy level of this particular beam corresponding to the energy of characteristic radiation of the element selected, and actually being slightly above that energy level to compensate for losses due to self-absorption and collimation, for example. In this sense, the arrangement employed for focusing of the beams is like that utilized in beta ray spectrometers of the magnetic semi-circular foscusing type which are utilized to control the impingement of beta rays from a source of radiation onto a photographic plate for measurement. In the present invention, suitable means are employed for analyzing the source of radiation such as the counter **20** and, in this connection, the actual apparatus used for ana-

lyzing and measuring the beta rays emitted from the source actually forms no part of the present invention and may be of the type described in detail in my copending application, Serial Number 586,172.

Broadly, another feature of the present invention is the method and means employed for introducing and removing each sample S from the evacuated space **16**. Essentially, the means devised represents a simple and highly practical way of advancing samples into the evacuated space in such a way as to overcome the necessity for re-establishing the necessary degree of vacuum for most accurate analysis after each sample is introduced, while preventing the introduction of gases from the sample itself. Thus, as generally designated by numeral **22**, in FIGURES 1–3, provision is made for a sample feeding system, to be described in detail, which is cooperative with the remainder of the apparatus to accomplish continuous analysis of a number of samples in succession with a minimum of adjustment and control and through utilization of a single source of energy notwithstanding the differences in energy level required for analyzing each element.

Now referring in more detail to the apparatus employed in the preferred form of my invention, the voltage-generating apparatus **10** may take the form of an electrostatic generator such as that devised by R. J. VandeGraaff; this may be characterized broadly as a high-potential generator capable of furnishing a steady voltage output. Briefly, and as illustrated, an electric charge is distributed onto a moving insulated belt **24** disposed in spaced relation within an insulating tube **25**, and one end of the belt projects into a first sphere **26** to receive an electric charge from a depositing brush **27**, which charge is conveyed along the moving belt into the center of a second sphere **28** having a pick-up brush **29**. By supplying the charge continuously at the center of the sphere, a high-voltage potential can be developed and, for example, a 14-inch sphere of polished aluminum would sustain about half a million volts. In addition, the maximum-limiting potential of the sphere is determined by the breakdown electric field in the gas at the surface of the sphere and, for example, in the presence of a small amount of pressure or through the use of a controlled atmosphere containing Freon, carbon tetrachloride vapor, etc., such would permit increasing the voltage to about a million volts on a sphere of reasonable size. As an alternative, double-strength Pyrex glass having an outside diameter of about ¾-inch may sustain a potential-voltage difference of about 500,000 volts; but in any event, the provision of half a million or a million volts represents a practical level from which a high-energy electron beam may be derived. One suitable electrostatic generating apparatus now commercially available is that made presently by the American Electrostatic Company and which is capable of delivering up to 20 microamperes of current. This current level corresponds with 1.25×10^{14} electrons per second which equals a radioactive strength of over 3,000 curies in terms of the amount of beta ray emitting material that would be required to produce the same number of electrons. In this connection, utilization of an electron beam is most favorable in the application of the present invention for the reason that the electrons produced by the bombardment resulting from high voltage can be directed on an extremely small spot, and in one direction, and the equipment itself is of a practical size for use either under field or laboratory conditions.

In order to accelerate the potential-energy charge developed in the sphere **28**, various discharge tube may be employed to accelerate this charge into a high-kinetic energy, concentrated electron beam. One type available and in general use for all types of electron generation is the hot cathode X-ray tube which permits utmost flexibility, adjustment, and maximum convenience in generating electrons which are to be directed against a target. However, in the practice of the present invention, where it is most desirable to have available a constant bombarding energy and to exclude other energies, it is greatly preferred to employ a gas-discharge tube which is capable of higher currents, more intense energy deliveries and more sharply defined beams. The gas-discharge tube is not tolerant of changes in applied voltage, but once adjusted, prefers to work at a particular fixed voltage and of course the apparatus **10** provides a constant applied voltage, so that it is possible to employ as the accelerating means, a cold cathode gas-discharge tube of electron accelerating tube, as illustrated at numeral **12**. Thus, as shown in FIGURE 1, the discharge tube **12** is comprised essentially of a pair of inner and outer concentric, heavy glass tubes **32** and **33** and wherein one end of the inner concentric tube projects through the wall of the sphere **28** and has a metal electrode **34** sealed to that end and spaced within the sphere. In order to conduct the charge from the sphere to the electrode, a pick-up wire **35** is shown extending therebetween. The opposite end of the tube **32** is similarly provided with a metal electrode **36** which is sealed to the glass and projects forwardly therefrom beyond the outer glass tube **33** and through an opening **37** formed in the evacuated chamber **16**. In the employment of this type of tube construction, no filament is required but merely through the utilization of electrodes at each end, the cathode ray beam is accelerated through the inner concentric tube and through a window such as an aluminum or beryllium window **38** at the end of electrode **36**, this window being of sufficient thickness to sustain the vacuum inside the tube at whatever level is desirable. Tubes of this nature and description have a tendency to evacuate themselves further in the course of being used, and it is common to provide some means for introducing traces of a gaseous material to maintain their condition. For example, a quantity of hydrogenized zirconium wire would serve excellently if provision to electrically heat it is afforded. As a residual gas, hydrogen is entirely suitable. Through this expedient, and by acceleration of a cathode ray beam through the tube **12**, there is made available within the vacuum chamber **16** a concentrated high-energy beam much above the energy level required for analysis of the specimen.

In order to conduct the analysis, the vacuum chamber **16** is defined by a generally rectangular enclosure formed of aluminum wall portions **40** and at ont end, a relatively thick glass window **41**. Positioned on the bottom wall **40** is a supporting post **42**, and to one side of the post above the window **38** a plate **44** is secured which includes a slit **45** aligned with the aluminum window **38**. Supported in spaced relation between the window **38** and plate **44** is the material absorber **14** which as shown may be secured to the plate **44**, and is disposed to intercept the concentrated beam directed through the discharge tube **12** for the purpose of scattering the beam into a plurality of particle rays of different energy levels, and substantially reducing the maximum energy below that of the original energy level.

Fundamentally, the material absorber **14** is selected to be of a thickness and area density which will subtract the least energy from the concentrated beam to provide a given maximum energy required for analysis of the specimens, and based on the given initial energy of the beam. Thus, for example, where a 500 kilovolt beam is to be reduced to an energy range having a maximum of 100 kilovolts, material such as beryllium and lithium may be selected for example where the thickness would be on the order of 0.010-inch. Electron beams passing through the material absorber will therefore have a fixed upper limit corresponding with the maximum limit of energy of individual electrons which get through the absorber and specifically, the flux of radiation through an absorber of the type described placed in the path of a 500 kilovolt beam, will contain a continuously-distributed population of electrons starting with very few in the close

vicinity of the maximum energy of 250 kilovolts, increasing in abundance for lower energies, passing through a maximum somewhere in the vicinity of 100 kilovolts and then becoming less abundant toward the very low energies. From this, it will be seen that by the simple expedient of placing a thickness of matter in the path of the radiation beam, there is created a concentrated source of electron radiation which has many of the characteristics of a kilocurie 250 kilovolt maximum beta ray emitter.

In that the flux of electrons emerging from the material absorber placed in the path of the concentrated beam is no longer unidirectional, but comes off in a variety of directions, it is then possible through utilization of a magnetic field passing through the vacuum chamber, perpendicular to the plane of the drawing, to cause the scattered radiation particles to become deviated and to transcribe a somewhat semi-circular path of travel through the magnetic field so as to impinge upon the plate **18**, as described. The plate **18** is suitably supported between the upright supporting posts **42** and a wall portion **47** with the aperture **19** positioned in desired alignment over the sample S. In addition, projecting upwardly from the supporting posts **42**, are a pair of spaced obstacles **48** which are provided with divergent openings **49** to selectively pass electrons deviated by the magnetic field in the desired energy range and to eliminate electrons of other energies outside the desired range so as to block their passage into the main analysis portion of the vacuum chamber.

Again, the general construction of the vacuum chamber for the purpose of properly focusing electrons of the desired energy level on the plate **18** closely follows the construction and arrangement of a magnetic, semi-circular focusing spectrometer and reference is made to the book entitled "Radiation from Radioactive Substances," by Rutherford, Chadwick and Ellis, FIGURE 94, page 32, 1930 edition. In accordance with known principles, as the magnetic field increases so does the energy of the electrons which are focused at a given point on the plate **18**. If, in the plate **18**, there is arbitrarily established an aperture at a particular point, and if the magnetic field parallel to the plate is made subject to control, then it is evident that one would be able to choose the energy of electrons which pass through the aperture merely by adjusting the magnetic field. Additionally, in the particular application of the present invention, it is desirable to position focusing electrodes **50** which are spaced on opposite sides of the path of travel of the electron beam passing through the aperture **19** as a supplemental means for accurately focusing electrons passing through the aperture in the form of a concentrated beam impinging on the specimen. A battery is illustrated at **52** electrically connected through wire **53** to the negative electrode, and a similar battery **52'** is illustrated as being connected through wire **53'** to the positive focusing electrode. The connecting wires **53** and **53'**, together with the connecting wires for the counter **20**, are secured in sealed relation to the vertical wall of the vacuum chamber by means of suitable metal field insulators, such as Stupakoff insulators **55** and such insulators may be employed throughout at the points of connection into the vacuum chamber.

To evacuate the entire chamber, a conduit **56** is led to a suitable vacuum pump and in accordance with conventional practice this connection may be served by a pumping device of considerable speed which would afford a controllable throttle to admit a very small amount of hydrogen elsewhere in the vessel, in order that the residual gas will correspond with material of the lowest possible density and beta ray absorbing power. As samples are brought into the space illustrated, and due to the improved nature of my sample feeding system, loading on the pump would be maintained at a minimum, and the amount of outgassing which a sample would exhibit presents no critical problem with a pump of reasonable size. Also, it is entirely feasible to use a reasonably small size for the entire structure in that at low energies, the electrons are very easily deviated and do not require strong magnetic fields at all. In fact, it is entirely suitable to use a pair of Helmholtz coils in the magnetic field to control very low energy electron beams. Also, it is desirable to avoid the use of iron in the magnetic field and thus avoid all complicated non-linear properties of the magnetic field that result from the presence of iron in it.

The improved apparatus devised for introducing samples into the vacuum space requires, as a preliminary consideration, knowledge of the desirable characteristic of grease as a means of permitting relative movement between contacting surfaces while resisting the intrusion of air at atmospheric pressure; and in fact, grease of a selected consistency would absolutely prevent such intrusion for an indefinite period of time. Particularly in thin layers, grease is characterized by the fact that the force available owing to difference in pressure, such as between the exterior and interior of the vacuum chamber, is not sufficient to produce a shearing stress to a degree that exceeds the strength of the grease itself. Grease also has the desirable characteristic of preventing any possible contamination of the interior evacuated space since, being a solid, it will resist movement of any contaminating or foreign particles into the sample space.

With the above in mind, the improved apparatus **22** of the present invention consists of a series of shallow depressions **60** located on the top horizontal surface **61** of a rotating block portion **62**, the latter being mounted for rotation on a spindle **63**. Each of the shallow depressions is in the shape of a right circular cylinder much shorter than its diameter, and preferably a series of four depressions is provided spaced at equal distances about the outer peripheral portion of the top surface **61**. The rotor itself is also of a generally cylindrical shape, composed of metal, with the axis of the cylinder aligned vertically and with the top surface of the cylinder cut off along a plane perpendicular to the axis for disposition of the depression **60** therein. As best seen from FIGURE 1, the rotor is arranged to slide within an inverted cup-shaped housing or cap **65** which is mounted in stationary relation at the lower right-hand portion of the vacuum chamber **16**, and this housing **65** is provided in its top surface with a series of four cap openings **66**, **67**, **68** and **69**, openings **66** and **67** being located exteriorly of the vacuum chamber **16** and openings **68** and **69** located interiorly thereof. Each of the openings **66**—**69** are of a size corresponding to the diameter of the shallow depressions **60** and are spaced a corresponding distance apart and with one opening **69** aligned centrally with the aperture **19** to provide access to a sample S contained in one of the depressions **60**.

Related to the housing or cap **65**, and adapted to separately serve each opening, is a schedule of functions repetitiously conducted in the following manner: Through cap opening **66**, a suitable vacuum sweeper arrangement **70** is diagrammatically represented in FIGURE 1 and is employed to sweep the bottom of the sample-carrying depression and to remove any dust which lays on it; through opening **67** and sealed to it, there is a dispenser **71** for depositing a small, fresh quantity of powdered sample into the center of the corresponding sample-carrying depression; through opening **68**, a vacuum connection **72** is provided which may extend to some conventional mechanical vacuum pump and which is capable of removing any residual gas from the powered sample and to partially evacuate the depression carrying the powdered sample. Opening **69** communicates with the interior of the vacuum space, as described, and when the sample-carrying depression is advanced into position in alignment with the space, it will be ready for analysis in the manner described. Briefly, therefore, the method of preparing each sample for introduction into the vacuum

chamber consists of the steps of first cleaning the sample container, depositing a new sample into the container, or depression, subjecting the container and sample to a vacuum operation and followed by performing the actual analysis through the method and apparatus described.

Of course, any suitable apparatus can be utilized for advancing the rotor and the samples contained in the depressions through 90° intervals for the various steps performed. In FIGURE 3, a drive mechanism 80 is illustrated as a typical example which broadly consists of means to advance the wheel 81 keyed to the rotor through each 90° interval with the sample-carrying depressions 60 aligned with each of the respective openings 66—69 in succession; and, following each quarter advancement, the wheel and rotor are stationary for a selected "dwell" period to permit each operation to be performed. For this purpose, the spindle 63 is shown keyed into a driven member consisting of a series of four bifurcated, radially-extending arms 82 on the wheel 81 which are spaced equally to correspond with the disposition of the depressions 60 and openings 66—69. The arms are interconnected through generally concave, webbed portions 83 which are symmetrically arranged with respect to the central axis of the driven member. A pinion 85 is employed as a driving member intermeshing with gear 86 and the latter gear includes on its top surface a cam member 87 together with a pin 88. The pin is formed to move into engagement with each of the bifurcated arms 82 as at A and to cause advancement of the driven member in response to rotation of the gears; as the pin moves out of engagement with each bifurcated arm as at B, the cam will move into engagement with the web portion 83, causing continued advancement of each arm to a position 90° removed from point A and so that the next arm will move into position at A. The pin 88 will continue its advancement during which time the arms 82 and the block 62 remain stationary, then will move into engagement with the next arm positioned 90° away from the arm advanced; as it moves into contact with the arm, it will continue its movement, carrying the arm 82 with it, to the point B. Thus, the driven member follows a continuous pattern of advancement over 90° intervals followed by intermittent stopping, which corresponds with the "dwell" periods necessary for the various operations to be performed on the sample prior to the introduction into the evacuated space.

As a possible alternative, it may be desirable to omit the rough vacuum step in which event a rotor with only three holes spaced at 100° may be employed. In this instance, the cycle would be the same as the above with the exception that one step would be emitted and the speed of pumping into the vacuum space where measurement is performed would be assumed to be high enough that sufficiently good vacuum would occur immediately after cleaning of the sample containers and holes. On the other hand, the question of whether or not a sufficiently good vacuum could be continuously maintained with a reasonable investment in pumping equipment must be resolved in selecting between a three or four position system.

In the following, there is set forth the manner in which sample measurement is conducted in accordance with the present invention, and the values applied are for the purpose of illustration, not by way of limitation. Fundamentally stated, measurements are conducted by determining the point of discontinuity or near approach to a discontinuity in the function of radiation output from the sample versus kinetic energy per particle for impinging electrons. Essentially, the method would therefore consist in oscillating the energy through a small increment, that is, oscillating the controlling magnetic field on the energy selector, for example, and the range of oscillation of the energy of the chosen particles would be such as to pass just above and below the *k* alpha absorption limit of the sample. The sample would be sensed by counter 20 which is adjusted to selectively gate the radiation corresponding with the *k* beta emission for the element about which data is being sought. The specific radiation would be clearly noted at the upper end of the chosen energy range of the impinging particles, and entirely absent at the lower end of the chosen energy range.

To focus the beam of electrons on the target or sample over a wide energy range, and as a preliminary to actual sample analysis, a determination should be first made as to the potential difference to be established across the electrodes 50 for each given energy level of electrons to be impinged upon the sample. The energy level of electrons passing through the aperture 19 will in turn be determined by the strength of the magnetic field and this may be regulated by controlling the current through the coils, not shown. As a specific example, a viewing window, also not shown, is positioned in the wall of the housing and a horizontal screen is placed at the lower end of the opening 69 to support a layer of zinc sulphide phosphor. The phosphor material may be of the type employed in making watch dials, or used as a coating on the inside of fluorescent lamps or television tubes. The current is then sent through the Helmholtz coils at a particular chosen value, such as for example, one ampere. It is emphasized at this point, however, that the current value to be chosen will depend upon the dimensions of the Helmholtz coils and upon the number of turns which they contain, and the designation of one ampere is entirely arbitrary and has no particular significance. At this level, electrons passing through the accelerator tube will be transmitted and scattered by the absorber 14 then passed by the apertures 45, 49 and 19, in succession, to impinge on the phosphorescent layer placed at "S" and create a spot of luminosity thereon. The luminous area can be adjusted by controlling the potential delivered on the electrodes 50 until a value is reached at which the luminosity is a concentrated narrow band of light seen as a strip which would appear perpendicular to the plane of the paper as shown by the phosphor placed at "S," all of which can be visually noted through the viewing window. Notations are then made of the coordinates of adjustment which produce the narrow strip of luminosity, so that for example the notation may appear as "one ampere—50 volts." Next, 1.05 amperes may be chosen for the current in the Helmholtz coils and the potential difference is again adjusted in order to establish a concentrated narrow band of light on the phosphorescent strip. Thus, a succeeding notation may be made, "1.05 amperes—53 volts." Correspondingly, the voltage values are given merely for the purpose of illustration and as the current values are incrementally increased over a wide range each potential difference value corresponding to the current value to establish the narrow strip of luminosity can be noted in order to produce a table derived from a succession of such determinations, so that for every given current value there will be a known voltage to bring about proper focusing of the electron beams. Of course, the table of values derived will be peculiar to each particular apparatus and the relative dimensions and spacing between elements. From the table, however, the variations involving the coils and the potential difference of electrodes 50 are reduced to a single variable so that it is only necessary to designate the current upon which the potential difference depends. Also, for the purpose of conducting an analytical procedure, it may be assumed that the accelerating voltage delivered by the static charge generator over the belt 24 onto electrode 28 is maintained at a relatively constant value by controlling belt speed to a relatively constant charge rate from the element 27. It may be assumed, also, that the choice of the obstacle 14 remains constant along with the size of the apertures and that all other elements of structure whereby electrons are produced and delivered into space

16 are held constant, permitting only changes in the current as the remaining variable.

In conducting the actual analyses of samples, each sample carrying depression is advanced successively into alignment with the opening 69 for analysis. As stated, the grease layer will be effective to exclude substantially all air and impurities from the chamber 16. In this way, minimum load is placed on the vacuum pumps and vacuum requirements are made less stringent due to the high intensity, concentrated beam made available for analysis. For each sample presented in the receptacle 69, the current in the Helmholtz coils is varied along with the potential difference setting across the electrodes 50 so that the electron beam will impinge on the specimen or sample. From this, the corresponding count-rate received in the counter 20 may be determined. Actually, this is indicated in the count-rate meter which is part of the system connected to the terminals 57, emerging from the geiger counter and passing out through the insulating terminal in the wall 40 of the space 16 and again, reference is made to my co-pending application for a description of the counting system. For a given sample, a stepwise experiment will consist of a tabulated set of values showing coil current as the independent variable and showing experimentally determined count-rate as the dependent variable and on a graph this would appear as a smoothly varying set of figures in the count-rate column except for the values of coil current just above and below certain analytically significant choices, the latter being related to specific emitting thresholds for electromagnetic radiation derived from the sample. Such emitting thresholds are extremely sharp and disturb the otherwise smooth variation of the figures in the count-rate column of the experimental table. The significance of this is twofold: First, the Helmholtz coil current corresponding with the last chosen value just below a discontinuity identifies the element in the sample which produced the discontinuity; secondly, the size of the discontinuity, that is, the difference in count-rate between the tabulated values just below the point where such a jump occurs and the first tabulated value where the variation becomes smooth again, will indicate the amount of the given element which is present in the sample. From the above, it will be seen that it is possible to recognize the element present and also to determine its amount. In this connection, the discontinuities appearing on the table are caused by the binding forces affecting electrons not participating in chemical linking, being inner electrons of the atoms of substances involved. Thus, for example, the percentage of calcium present in a sample is determined with no reference to whether the calcium is present as carbonate, sulphate, silicate, or other chemical forms.

From the above, it may be broadly stated that the discontinuities will be found at values for which $H\rho$ of the electrons focused on the sample are in the close vicinity of the excitation potentials for the inner shells of the electrons of the atoms which are being recognized in the sample, $H\rho$ being a quantity designating the product of the magnetic field and the radius of curvature of an electron of a given kinetic energy. This value is constant for an electron of any given energy and for each value there is an exactly corresponding number of electron volts for electons which will exhibit this characteristic. As a result, the discontinuity will be plotted in values which are proportional to $H\rho$, since the geometry of the apparatus will determine ρ and the coil current will determine H. To graphically analyze the results, the coil current versus the potential difference at the electrodes 50 are made to be dependent on a third variable, time. So, a recorder which has a uniform progression of its chart paper with respect to time can be connected to the output of the count-rate metering circuit which at every instant produces an electric current delivered to the input recorder in a manner to actuate the pen in the direction of the abscissa of the coordinates. Accordingly, and as stated, the discontinuity will be plotted in values proportional to H and will increase uniformly with time as the current values are correspondingly increased.

It is understood that, as in the previous non-automatically recorded example, attention is paid to the providing of necessary values of focusing potential on the electrodes 50. This may be done by using a suitably shaped cam, through which, by means of a rack and pinion, or some other suitable arrangement, the polar graph represented by the cam acts upon the rotatable shaft of a suitable potentiometer. Assuming that the value of H and the corresponding current required to produce it are planned to increase at a uniform rate with respect to time, the cam arrangement which is situated on a shaft (connected by solid gearing to the same drive that controls the increase of the coil current) will properly control and relate the focusing potential, delivering at every instant a suitable focusing potential on the electrodes 50, to correspond with the coil current which exists at that instant.

From the foregoing, it will be evident that there has been provided a highly efficient, practical and simplified way of analyzing various substances and compounds to determine the amount of selected elements therein. Here, the improved method and apparatus of the present invention is capable of exciting the characteristic radiation of any selected element by furnishing the necessary high-intensity energy in the form of a concentrated particle beam which is focused on a sample introduced in a unique way into an evacuated space. The entire method and apparatus is espectially unique by virtue of its ability to excite characteristic radiation from low atomic number elements through the use of a particle stream of electrons; but also, the particle stream of electrons furnished for excitation may be simply and accurately controlled to be of the necessary resonant efficiency level for analysis of virtually any element contained in the sample. As mentioned, this is brought about through the provision for a source of high-energy electrons in the form of a concentrated beam which may be diffused over an enrgy range, including the energy levels required to excite characteristic radiation of the known elements and then the energy level required for characteristic radiation of each element to be analyzed may be selectively focused under the influence of a magnetic field of controllable strength to bombard a sample containing the known element. Through conventional means, the characteristic emission of radiation from the sample may be detected to determine the amount of known element present in the sample. By developing a high-intensity, concentrated beam which may be accurately focused on a very limited area of the sample, vacuum requirements for the analysis space are not so absolute and may be termed more as a partial vacuum. Here, the term "partial vacuum" has reference to a pressure of one ten thousandth of an atmosphere, with residual gas, hydrogen. In considering vacuum requirements, the sample feeding system of the present invention also reduces the pumping requirements for maintaining the desired vacuum level due to the unique manner in which the samples may be rapidly and successively advanced into the chamber. A further practical advantage, particularly in field use, is that the entire apparatus is relatively compact and permits the use of commercially available and relatively inexpensive equipment.

It is to be understood further from the foregoing that various changes and modifications may be made in accordance with the present invention, especially in the particular type of apparatus employed, without departing from the scope of this invention as defined by the appended claims.

What is claimed is:

1. In apparatus for conducting analysis of known elements in a substance disposed in an evacuated space with

means provided for detecting characteristic emission from the element, the combination comprising a high voltage source of electrons, means extending between the high voltage source and the evacuated space for acceleration of the electrons into the form of a high kinetic energy concentrated beam for discharge at one end into the evacuated space, a material absorber located at the discharge end of said accelerating means to scatter the beam into particle streams of electrons, a magnetic field passing across the evacuated space to direct the particle streams toward the substance, a plate including an aperture aligned with the substance to intercept the particle streams, said magnetic field being of a controllable strength to selectively direct electrons in the particle stream corresponding to the energy of characteristic radiation of the element on the aperture for impingement on the substance whereby to exite characteristic radiation of the element.

2. In apparatus according to claim 1 in which a static charge generating apparatus is employed to provide the high voltage source of electrons at a substantially constant applied voltage.

3. In apparatus according to claim 2 in which a cathode discharge tube is provided for acceleration of the electrons into the evacuated space.

4. In apparatus according to claim 1 in which focusing electrodes are interposed between said plate and the substance to be analyzed.

5. In apparatus for conducting analysis of known elements in a sample disposed in upwardly facing position in an evacuated space with means for detecting characteristic emission from the elements, the combination comprising a high voltage source of electrons, an electron accelerating tube extending between the high voltage source and the evacuated space for acceleration of the electrons into the form of a high kinetic energy concentrated beam for discharge at one end of the tube vertically upward into the evacuated space, a material absorber located in spaced relation to the discharge end of said accelerating means to scatter the beam into particle streams of electrons of a predetermined maximum energy level, a magnetic field passing across the evacuated space to direct the particle streams along a generally semi-circular path toward the sample, a plate including an aperture aligned with the sample to intercept the particle streams, said magnetic field being of a controllable strength to selectively direct electrons in the particle stream corresponding to the energy of characteristic radiation of the element on the aperture for impingement on the sample whereby to excite characteristic radiation of an element for detection.

6. In apparatus for conducting analysis of known elements in samples disposed in an evacuated space with means for detecting characteristic X-ray emission from the element, the combination comprising static charge generating means for producing a high, constant voltage source of electrons; a cold cathode discharge tube extending between said generating means and the evacuated space for acceleration of the electrons into the form of a high kinetic energy concentrated beam for discharge at one end of said tube into the evacuated space, a material absorber located at the discharge end of said accelerating means being of an areal density to scatter the beam into particle streams of electrons over an energy range covering the energies of characteristic radiation of the elements to be analyzed, a magnetic field passing across the evacuated space to direct the particle streams along a generally semi-circular path toward the sample, a plate including an aperture aligned with the sample to intercept the particle streams, said magnetic field being of a controllable strength to selectively direct electrons in the particle stream corresponding to the energy of characteristic radiation of the element to be analyzed for impingement on the sample and focusing means interposed between said plate and the sample for establishing a potential difference in voltage across the path of electron flow according to the strength of the magnetic field to focus the electrons in the form of a concentrated beam on the sample whereby to excite characteristic radiation of that element for detection.

7. Apparatus for exciting characteristic radiation of known elements for analysis of samples containing the elements wherein there is provided an evacuation chamber to receive each specimen together with means associated with the chamber for detecting characteristic emission from each specimen, the combination comprising sample feed means associated with the evacuation chamber being adapted to receive selected samples outside the chamber containing the elements to be analyzed for evacuation and advancement into the interior of the chamber for analysis and with means for intermittently advancing and stopping said feed means to align each sample in succession for analysis within the chamber; means for developing a high voltage source of electrons, an electron accelerating tube extending between the source of electrons and the lower end of said evacuation chamber for accelerating the electrons into a high kinetic energy concentrated beam for discharge from one end of said tube into the evacuation chamber, a material absorber positioned within the chamber to scatter the concentrated beam into particle streams of electrons over a predetermined energy range having a maximum energy corresponding to the maximum energy of characteristic radiation of the elements to be analyzed and with the path of the concentrated beam into the evacuation chamber being parallel but spaced in relation to the exposed surface of the samples containing the element to be analyzed, a plate positioned to extend horizontally in vertical spaced relation above the sample including an aperture therein aligned directly above the sample, a magnetic field passing across the evacuated space to direct the particle streams along a generally semi-circular path striking said plate, the strength of the magnetic field being adjustable to selectively focus electrons in the particle stream, of an energy corresponding to the energy of characteristic radiation of the element on the aperture, and focusing electrodes interposed between said plate and said housing to direct the electrons passing through the aperture for impingement on each sample brought into alignment with the aperture to excite the characteristic radiation of a known element in each sample for detection thereof.

8. Apparatus for exciting characteristic radiation of known elements for analysis of specimens containing the elements wherein there is provided an evacuation chamber to receive each specimen together with means associated with the chamber for detecting characteristic emission from each specimen, the combination comprising a housing projecting partially into the chamber providing a series of upwardly-facing openings with at least one opening being located within the chamber and communicating with the interior of the chamber, means rotatable in air-tight relation within the housing and having a series of specimen-carrying depressions corresponding in diameter and spacing with the openings in the housing, and means for intermittently advancing and stopping said rotatable means to align each depression in succession with an opening in the housing and provide for successive depositing, evacuation, and movement of each specimen into the interior of the chamber for analysis; means for developing a high voltage source of electrons, a cathode discharge tube extending between said apparatus and the lower end of said evacuation chamber for accelerating the high-voltage source of electrons into a high kinetic energy concentrated beam for discharge from one end of said tube into the evacuation chamber, a material absorber positioned within the chamber to scatter the concentrated beam into particle streams of electrons over a predetermined energy range having a maximum energy corresponding to the maximum energy of characteristic

radiation of the elements to be analyzed and with the path of the concentrated beam into the evacuation chamber being parallel but spaced in relation to the exposed surface of the specimen containing the element to be analyzed, a plate being positioned to extend horizontally in vertical spaced relation above the specimen including an aperture therein aligned directly above the specimen, a magnetic field passing across the evacuated space to direct the particle streams along a generally semi-circular path striking said plate, and the strength of the magnetic field being adjustable to selectively focus electrons of an energy corresponding to the energy of characteristic radiation of each element to be detected on the aperture for impingement on each specimen.

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