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Packard et al.

[11] **3,898,457**

[45] **Aug. 5, 1975**

[54] **METHODS AND APPARATUS FOR HANDLING FLEXIBLE LIQUID SAMPLE CONTAINERS FOR SCINTILLATION SPECTROMETRY**

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[73] Assignee: **Packard Instrument Company, Inc.**, Downers Grove, Ill.

[22] Filed: **Nov. 1, 1973**

[21] Appl. No.: **411,839**

Related U.S. Application Data

[63] Continuation of Ser. No. 192,543, Oct. 26, 1971, abandoned.

[52] **U.S. Cl.** ..... **250/303**; 23/253 R; 250/367; 250/494

[51] **Int. Cl.** **G01t 1/204**

[58] **Field of Search** ..... 23/253 R; 250/303, 304, 250/252, 366, 367, 494, 496, 497, 302

[56] **References Cited**

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2,750,514	6/1956	Armistead	250/366
3,526,480	9/1970	Findl et al.	23/253 R
3,540,858	11/1970	Rochte et al.	23/253 R
3,620,678	11/1971	Guigan	23/253 R
3,688,120	8/1972	Packard	250/252
3,713,779	1/1973	Sirago	23/253 R

Primary Examiner—Harold A. Dixon

Attorney, Agent, or Firm—Wolfe, Hubbard, Leydig, Voit & Osann, Ltd.

[57] **ABSTRACT**

Methods and apparatus for handling flexible sample containers used for liquid scintillation spectrometry analysis where detector photomultiplier tubes are moved axially together "squeezing" the flexible container therebetween in the counting station to achieve close optical coupling. The sample containers may be made up of layered, flexible, light-transmissive, polyester film wherein the sample container portion is defined by joining the film layers. The flexible container may be individual units or a continuous strip and may include provision for identification data or records by way of a magnetic strip or punch character designation. Optimum counting efficiency is achieved with round flask-like configurations where the flexible container conforms to the shape of the photomultiplier tubes used therewith.

8 Claims, 9 Drawing Figures

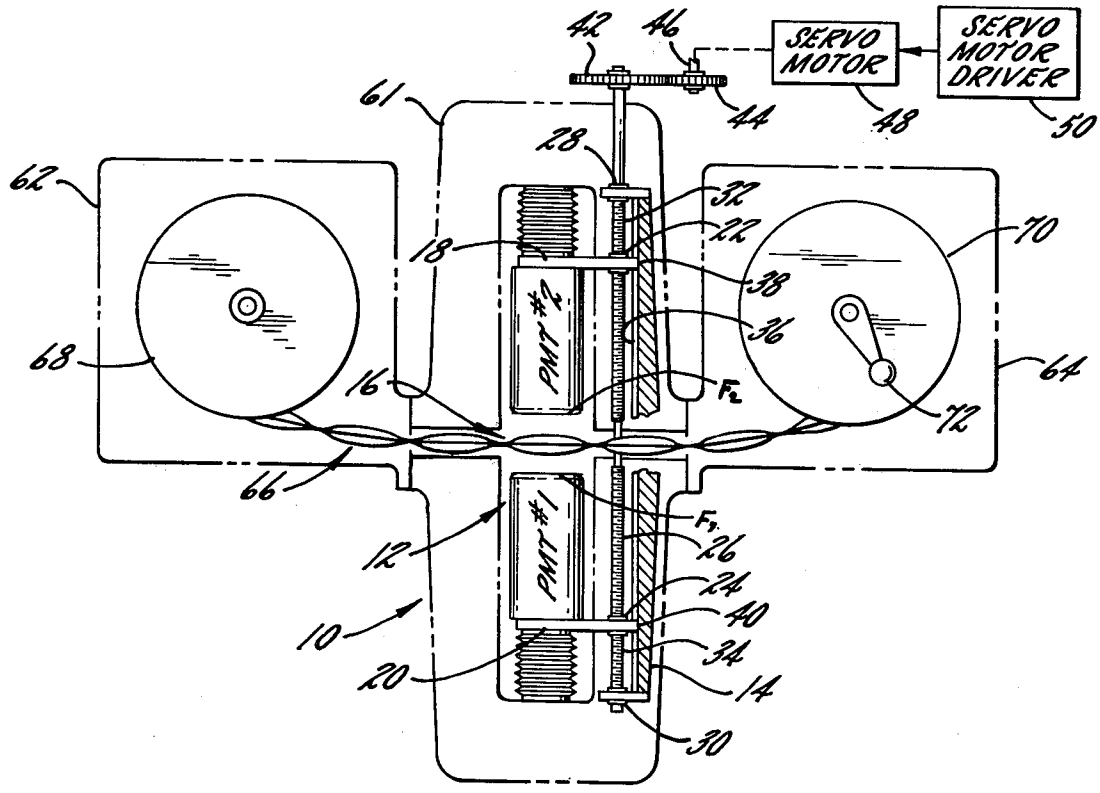
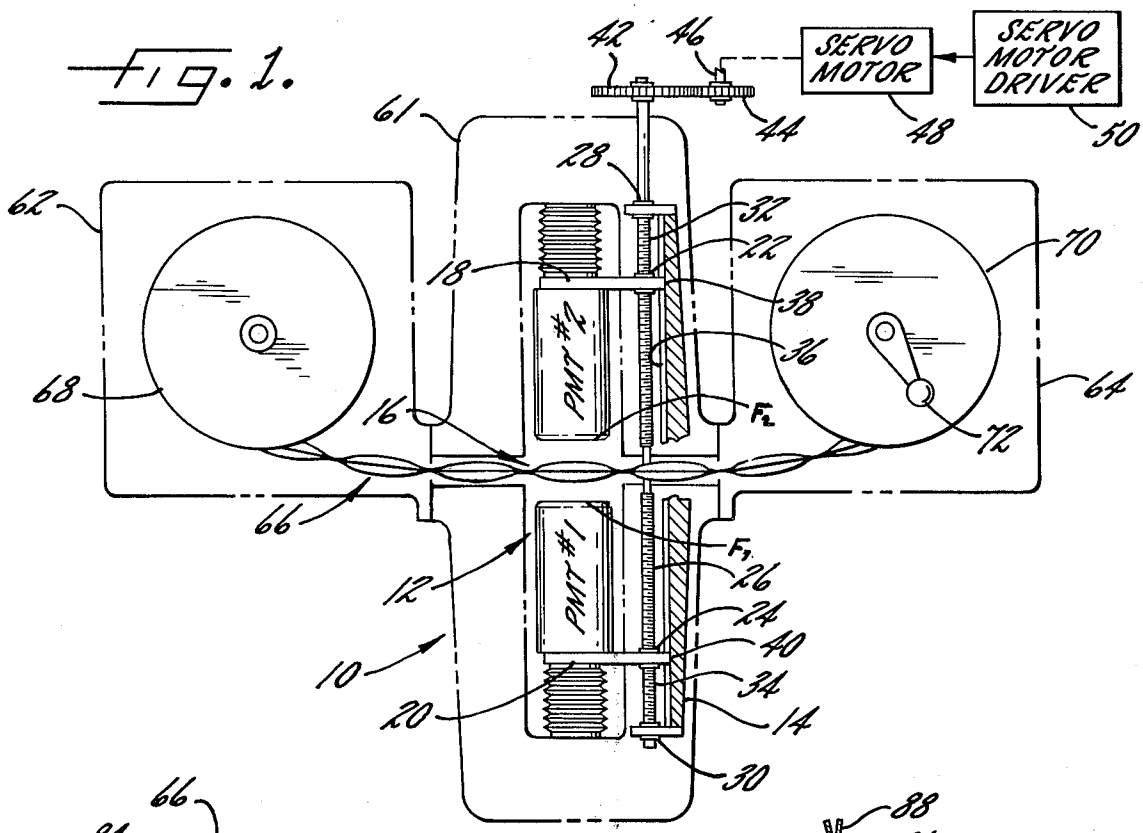


Fig. 1.



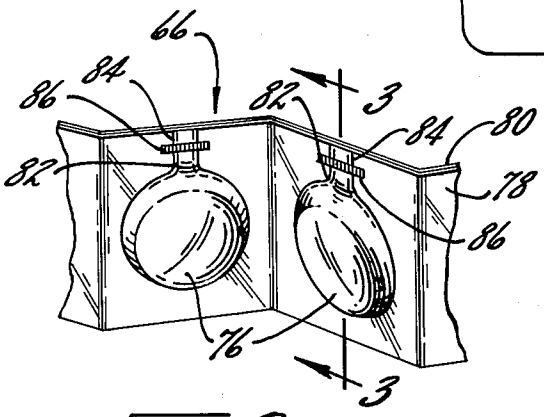


Fig. 2.

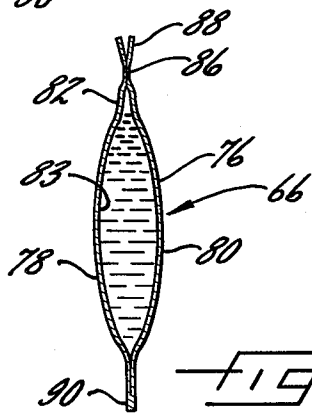


Fig. 3.

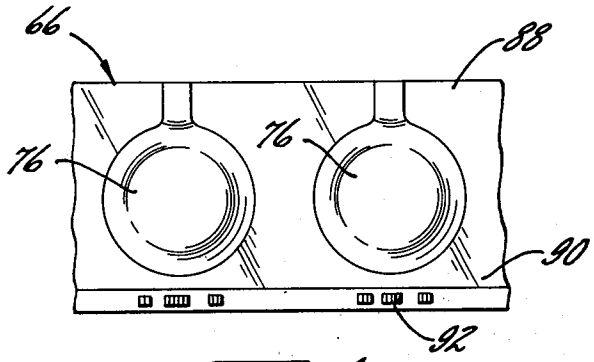


Fig. 4.

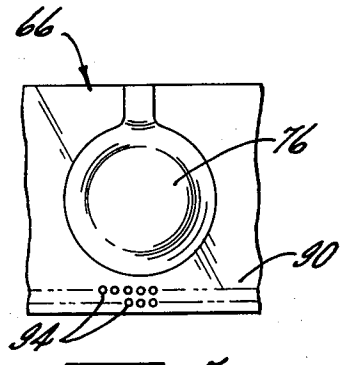


Fig. 5.

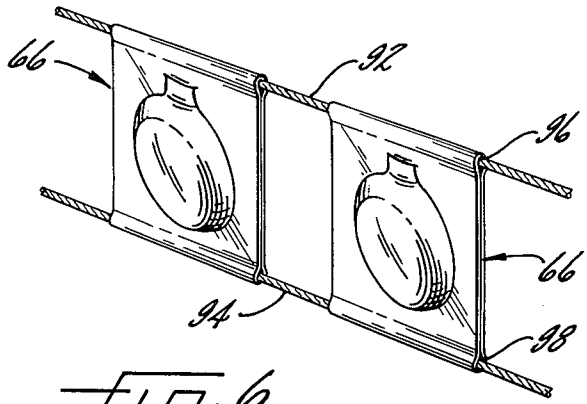


Fig. 6.

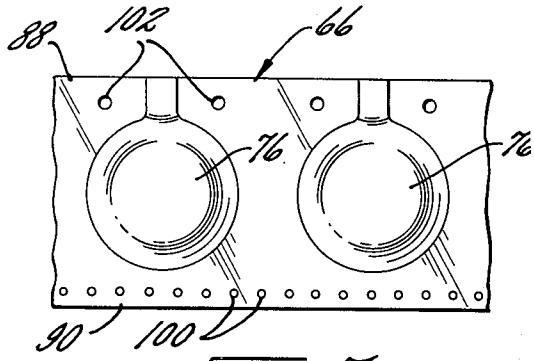


Fig. 7.

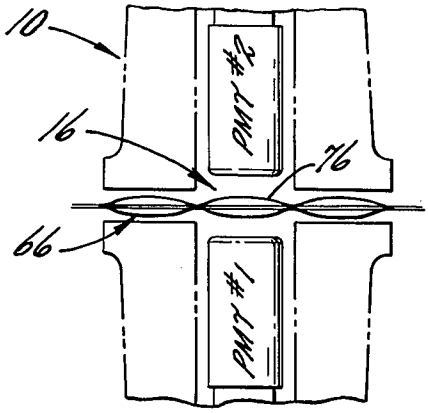


Fig. 8.

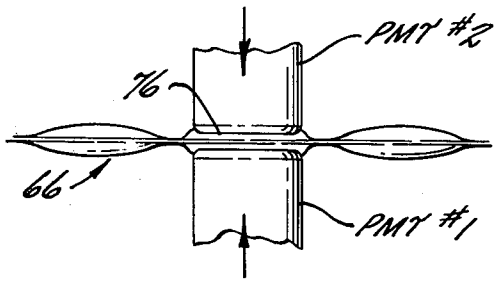


Fig. 9.

METHODS AND APPARATUS FOR HANDLING FLEXIBLE LIQUID SAMPLE CONTAINERS FOR SCINTILLATION SPECTROMETRY

This is a continuation of application Ser. No. 192543 filed Oct. 26, 1971 and now abandoned.

RELATED APPLICATIONS

Lyle E. Packard Application Ser. No. 630,892 filed Apr. 19, 1967, assigned to the assignee of the present application, and now U.S. Pat. No. 3,688,120.

DESCRIPTION OF THE INVENTION

This invention relates generally to methods and apparatus for handling sample containers for liquid scintillation spectral analysis of test samples containing one or more radioactive isotopes disposed in a liquid scintillator. More particularly, the invention relates methods and apparatus for handling the flexible sample containers for liquid scintillation detection operations in a wholly automatic manner, yet which permit maintainance of high counting efficiency and quantitative accuracy of result.

Background of the Invention

In conventional liquid scintillation counting, a radioactive nuclide dissolved or dispersed in a scintillator liquid is contained in a thin plastic or glass sample vial. Liquid scintillation spectrometers, or apparatus designed to provide spectral analysis of the test samples contained in such vials have been successfully utilized now for several years. One system which has found great acceptance today by people employing such sophisticated equipment is shown in U.S. Pat. No. 3,188,468 to L. E. Packard, assigned to the assignee of the present invention. Such apparatus is loaded with trays holding sample vials containing the scintillator and isotope, and the vials are manipulated seriatim into and out of a photomultiplier detection mechanism.

One of the important determinants of the quantitative accuracy of scintillation detectors is the efficiency with which the photomultiplier tubes can detect the scintillations. Some light is inevitably lost at the surfaces of the sample vial and of the photomultipliers, and as a consequence the efficiency of light detection is reduced. In other words, the closeness of optical coupling is one of the factors entering into an efficient scintillation counting apparatus.

It has been recognized that close optical coupling is desirable for high counting efficiency. In the aforementioned Packard U.S. Pat. No. 3,188,468 close optical coupling is achieved with the use of "light pipes" which are blocks of plastic having a central vial-receiving well, and with all external surfaces other than those facing the photomultipliers coated with a reflective material. Other arrangements for achieving the close optical coupling have been suggested such as vials having flexible walls adjacent the sides facing the photomultipliers and which walls conform to the surface of the photomultiplier tubes so as to remove air gaps. One such arrangement is illustrated in U.S. Pat. No. 2,750,514. Also, close coupling has been achieved with the use of a liquid having an index of refraction similar to that of the glass or plastic vial.

Another technique employed for improving counting efficiency has been described in an article by Gopi N. Gupta, "Simplified Solid-State Scintillation Counting on Glass Microfilm Medium in Plastic Bag for Hydrogen-3, Carbon-14 and Chlorine-36 in Biological and Organic Material" which was published in Anal. Chem., 39, 1911 (1967), The author used a heat sealed polyester film bag to contain a radionuclide-impregnated fiberglass sheet, the bag in turn being placed within a conventional plastic counting vial.

Accordingly, it is a general aim of the present invention to provide improved flexible sample containers handling methods and mechanisms for liquid scintillation spectrometry characterized in that the containers may be constructed at relatively low cost and can be easily filled, handled and subjected to spectral analysis with optimum counting efficiency.

A more detailed objective of the invention is the provision of a flexible sample container which is capable of being mechanically handled, and eliminates the need for extraneous vials, external light pipes or the like and yet is conformable with the surface(s) of the photomultiplier tube(s) to optimize counting geometry.

It is still another object of the present invention to provide a flexible sample container which may be formed in a continuous strip wherein a plurality of samples may be stored and handled in a compact assembly.

Yet another objective of the invention is the provision of handling, changing and transfer mechanisms which will operate with a continuous strip of flexible sample containers of the foregoing type.

DESCRIPTION OF DRAWINGS

Other objects and advantages of the invention will become apparent upon reading the following detailed description and upon reference to the drawings in which:

FIG. **1** is a front sectional view of an exemplary radioactive sample handling and measuring apparatus, housed in a suitable cabinet and depicting one form of handling of flexible containers of the present invention;

FIG. **2** is an enlarged view of one form of a portion of a strip of flexible containers of the present invention used in conjunction with the apparatus shown in FIG. **1**;

FIG. **3** is an enlarged view of a filled flexible container of the type shown in FIG. **2**;

FIG. **4** is an enlarged, fragmentary view of another form of a strip of flexible containers incorporating the features of data recording and identification of the present invention;

FIG. **5** is an enlarged fragmentary view of a flexible container incorporating another form of data recording and identification;

FIG. **6** shows another version of a strip of flexible containers, here illustrating individual containers attached to conveyor strips;

FIG. **7** is an enlarged fragmentary view of a further form of a strip of flexible containers, here depicting sprocket and locating aperatures provided along the marginal edges thereof;

FIG. **8** is a diagramatic view illustrating a strip of flexible containers moving in position between spaced apart photomultiplier tubes of the detector apparatus for introducing a sample container therebetween; and,

FIG. **9** is a diagramatic view similar to FIG. **8**, here depicting the position of the flexible container with respect to the photomultiplier tubes during counting.

While the invention is susceptible of various modifications and alternative forms, specific embodiments thereof have been shown by way of example in the

drawings and will herein be described in detail. It should be understood, however, that it is not intended to limit the invention to the particular forms disclosed, but, on the contrary, the intention is to cover all modifications, equivalents and alternatives falling within the spirit and scope of the invention as expressed in the appended claims.

General Apparatus Organization

Turning first to FIG. 1, there is shown a liquid scintillation apparatus, indicated generally at **10**, which houses a scintillation detector mechanism or apparatus **12**. For details of a detection apparatus generally similar to the one depicted in FIG. 1, cross reference is made to copending application Serial No. 630,892 filed Apr. 14, 1967 of Lyle E. Packard, which although serving a purpose different from that of the present invention, provides an arrangement of components as to the detection mechanism which is suitable for use in accordance with the present invention. Thus, the detection apparatus **12**, as viewed in FIG. 1, includes a base assembly **14** which houses a pair of photomultipliers PMT No. 1, PMT No. 2 disposed on opposite sides of a counting station **16**.

The photomultipliers PMT No. 1, PMT No. 2 are mounted so as to permit controlled simultaneous movement toward and away from the counting station **16**. To accomplish this, in the exemplary arrangement the photomultipliers PMT No. 1, PMT No. 2 are respectively mounted on depending brackets **18,20**, the latter having coaxially aligned, non-rotatable, internal oppositely-threaded bearing sleeves **22,24** rigidly mounted therein. A parallely disposed actuating shaft **26** passes through the aligned bearing sleeves **22,24** and is supported at one end in a bearing assembly **28** mounted on the base assembly **14**, and near its opposite end in a similar bearing assembly **30** also rigid with the base assembly **14**.

The end of the shaft adjacent the photomultiplier PMT No. 1 is provided with a left hand threaded portion **32** threadbly engaging with the bearing assembly **28** and bracket **18** supporting photomultiplier PMT No. 1. The other portion of the shaft **26** is provided with a right hand threaded portion **34** which is threadably engaged with the bearing sleeve **24** and bracket **20** supporting the photomultiplier PMT No. 2. The two brackets **18,20** and hence the photomultiplier tubes are maintained free for controlled movement toward and away from one another along a common axis while being prevented from rotating about the axis of the actuating shaft **26** by provision of a longitudinally extending track **36** fixedly mounted on the base assembly **14** and slidably engaged in complimentary shaped grooves **38,40** formed in the brackets **18,20**.

In order to enable rotation of the actuating shaft **26**, and hence movement of the photomultiplier tubes linearly toward and away from one another, one end of the actuating shaft portion **32** projects slightly beyond its bearing support **28** and is drivingly coupled to a gear train **42,44** (here shown only in diagramatic form, it being understood that the particular means employed for driving shaft **26** could take any of well known conventional forms and could, if desired, be mounted within the base assembly **14**). Moreover, a yieldable mounting may be employed to limit the movement of the tubes toward each other on the basis of reaction forces applied thereto. The movement of the photomultiplier tubes are more fully discussed below permits close optical coupling with the sample containers.

As illustrated in FIG. 1 the gear **44** is coupled to the drive shaft **46** of a conventional reversible servo motor **48** which receives its input from a servo motor drive **50**, the latter being provided with a control signal derived from a control module (not shown).

Pursuant to the invention, the housing **61** (shown in phantom) is provided with a sealed sample source supply chamber **62** adjacent the input side of the detector and a sealed sample receiving chamber **64** attached to the housing **61** adjacent the output side of the detector. A plurality of sample containers indicated generally at **66** are carried by a reel **68** in the supply chamber **62** and are transferred in seriatim order through the counting or detecting station **16** and then to a receiving reel **70** in chamber **64**.

In order to move the sample containers from reel **68** to reel **70**, the latter is provided with a crank handle **72**. However, it will be appreciated that a suitable motor and control may be utilized for automatic operation without departing from the invention.

Flexible Container Arrangement

In accordance with the present invention, the sample container is in the form of a flexible bag made up of a layered, light-transmissive polyester film which is heat sealed, welded or otherwise suitably joined to define a closed flask between the layers. To this end, referring to FIG. 2 there is shown in prespective a portion of a strip of flexible containers **66** particularly suitable for use with the foregoing described detector apparatus **10**. The individual flexible sample containers **66** are produced from a pair of strips **78,80** of polyester film such as polyethylene terephthalate (Mylar) or any other suitable film materials that are available to those skilled in the art. The centrally disposed flask-like portion **76** is formed by joining the layers **78,80** around its periphery, and it is preferably circular in the plane of the layers with a diameter closely approximating the diameters of the photomultiplier tubes. This arrangement enables the flask portions to conform with the round photomultiplier tubes and provides the optimum counting geometry for the container structure.

In order to enable filling of the flask portions **76** with test samples, the joining of the layers in the present instance is provided with a discontinuity **82** that permits the injection of the test samples into the centally located closed chamber **83** of the flasks. To further facilitate filling operations, the joint line defining the flask portion periphery may be brought up along vertical or angular paths to the top of the strips so as to form necks **84**. After the flasks have been filled with the test samples, sealing to prevent escape of the sample may be readily accomplished by a transverse joint **86** across the neck **84** defining lines. Of course, the joint may be made continuous in the first instance and broken open for filling followed by resealing. On the other hand, self-healing material may be employed to enable filling by a hollow needle or the like.

A filled container **66**, FIG. 3 assumes a generally pillow-shaped configuration, viewed edgewise, wherein the side walls become convex when the flask is as completely filled with liquid as possible. The width of the strips **78,80** are preferably larger than the diameter of flask portion **76** formed therein so that top and bottom

borders **88,90** respectively are left along the entire strip of containers.

In accordance with one of the features of the present invention, provision is made for the container strips to carry identifying data or information which may be mechanically, magnetically, optically or electrically handled for use. To this end, referring to FIG. **4**, there is illustrated one form of information carrying arrangement for a flexible container strip **66** wherein the lower border **90** is provided with a magnetic data recording strip **92**. The magnetic strip direcly beneath the flask portions is sufficient to carry a recorded identification of the sample program instructions and data for operating the counting apparatus and may also be used to store any results obtained with the scintillation counting apparatus.

Referring now to FIG. **5** there is illustrated another form of information or identification carrying arrangement rendered possible with the flexible container construction of the present invention. Thus, it is here shown that the lower border **90** of the flexible container strip **66** is provided with punched code designations **94** which may be utilized with conventional and known types of punched tape readers. Since the punched characters of the arrangement of FIG. **5** as well as the magnetic strip arrangement of FIG. **4** are integral with the flexible containers there is little possibility that the data will be lost or misplaced.

Flexible Container Transference and Handling

In accordance with another feature of the present invention, the provision of the flexible containers in a continuous strip enables a simplified handling in the detection apparatus and eliminates the need for much of the sample handling and transfer equipment which was previously utilized with individual sample vials. As shown in FIG. **1**, the strip of flexible containers may be compactly stored on a reel and the transference of the samples through the detector apparatus involves a passing of the strip from reel to reel.

Moreover, the flexible containers may be produced individually or, after production in a continuous strip, they may be severed into individual units. When such individual units are produced, the individual units may be attached to a conveyor strip or the like that is adapted to pull them in sequential order through the detector apparatus. An arrangement of that type is shown in FIG. **6** where a pair of wire-like elements **92,94** passing through sleeves **96,98,** respectively, formed at the top and bottom borders of the flexible containers **66** carrying individual containers in the same manner as would a continuous strip.

As an alternative mechanical means for transferring a flexible container **66** of the present invention through a detector apparatus, there is shown in FIG. **7** an arrangement where a plurality of spaced sprocket holes **100** are disposed along the lower border **90** of a container strip which in the present instance is shown with a pair of containers **76**. The arrangement is such that conventional strocket wheels or a belt with correspondingly spaced pins may be utilized to pull the flexible containers through the counting station **16** of a detection apparatus such as that viewed in FIG. **1**. In addition, as shown in FIG. **7** pilot holes **102** may be disposed at the top border **88** of the strip to serve as locaters that insure precise orienting of the flask portion **76** between the photomultiplier tubes of the detection apparatus.

Detector Apparatus

Pursuant to the invention, close optical coupling with the flexible containers **66** is achieved by "squeezing" contact between the photomultiplier tubes and the flask portions **76** of the containers. According to the invention, this may be accomplished by axially shifting the photomultipliers with respect to a container in the counting station. To this end, referring to FIG. **8**, and FIG. **1** conjointly, it is seen that the flexible containers carrying the test samples to be counted are brought into the counting station **16** of detector apparatus **10** in seriatim order. During the time when the containers are being shifted into and out from the counting station, the photomultiplier tubes PMT No. **1** and PMT No. **2** are in the retracted, axially spaced apart position. When the flexible container **66** holding the test sample to be analyzed is in the counting station **16**, the photomultiplier tubes are then moved axially together until they are brought into intimate contact with the outer faces of the flask portion **76**, as shown in FIG. **9**.

The photomultiplier tubes are brought together with sufficient pressure so as to "squeeze" the flexible container **76** causing the outer walls of the flask portion to conform with the faces of the photomultiplier tubes. This results in an extremely close optical coupling between the tubes and the container which minimizes lost light at the contact surfaces of the container and tubes and high quantitative accuracy of liquid scintillation counting results.

Of course, it will be appreciated that a similar result may be achieved by maintaining the photomultiplier tubes in a fixed spaced apart axial position and pulling the container into position in the counting station between the faces of the tubes. If the spaced apart distance between the tubes is preselected so as to be slightly less than the thickness of the filled containers, the squeezing will result from the pulling of the container between the tube faces.

We claim as our invention:

1. In an apparatus for spectrally analyzing test samples containing one or more radioactive isotopes disposed in a liquid scintillator and adapted to handle a plurality of sample containers, the combination comprising, housing means including a source of said sample containers, at least one photomultiplier detection device and storage means for receiving said containers, said photomultiplier detection device being disposed adjacent a linear path extending between said sample source and said sample storage means, a light-tight counting station between said sample source and said sample storage means and disposed on said path, means for conveying said sample containers in a continuous strip from said source along said path and individually into the counting station and, thereafter, into said storage means, means for axially shifting said photomultiplier detection device toward and away from the counting station, and a second photomultiplier detection device in a longitudinally spaced apart relation to said other photomultiplier detection device.

2. Apparatus as claimed in claim **1** wherein said conveying means simultaneously transfers adjacent ones of said containers from said source to said counting station and said storage means.

3. In an apparatus for spectrally analyzing test samples containing one or more radioactive isotopes disposed in a liquid scintillator and adapted to handle flexible sample containers formed by joining layered, flexible, light-transmissive, polyester film to define a bag portion therein, the combination comprising, housing means including a source of said flexible sample containers, at least one photomultiplier detection device and storage means for receiving said flexible containers, said photomultiplier detection device being disposed between said sample source and said sample storage means, a light-tight counting station adjacent said photomultiplier detection device, means for axially shifting said photomultiplier detection device toward and away from said counting station so that said flexible containers are squeezed against said photomultiplier detection device during said spectrally analyzing, means for conveying said flexible sample containers from said source into the counting station and thereafter into said storage means, and said flexible containers are disposed on a continuous strip and said conveying means simultaneously transfers adjacent ones of said flexible containers from said source to said counting station and said storage means.

4. A method of spectrally analyzing test samples containing one or more radioactive isotopes disposed in a liquid scintillator and confined in a sample container, comprising the steps of, storing a plurality of said containers at one side of a counting station having at least one photomultiplier detection device adjacent said counting station, shifting said containers in a continuous strip from said source to the counting station, locating individual ones of said containers of the strip in the light-tight counting station, counting the scintillations produced within the sample container in the counting station, shifting the counted sample container out of said counting station to a storage means at the opposite side of said counting station, said sample container being one of a plurality of flexible containers formed of layered, light-transmissive, polyester film, spacing said photomultiplier detection device during said counting at a distance from a surface adjacent the counting station which is less than the thickness of the filled sample containers such that said containers when shifted into the counting station are squeezed between said photomultiplier detection device and said surface so that the face of the container in the counting station conforms to the face of the photomultiplier detection device.

5. A method as claimed in claim 4 wherein said surface comprises another photomultiplier detection device.

6. A method as claimed in claim 4 wherein said container is formed by joining at least a pair of layers of flexible light-transmissive polyester film and conforms in geometry to the contacting surface of said photomultiplier detection device.

7. A method as claimed in claim 4 wherein said spacing step includes axially shifting said photomultiplier detection device toward said sample container in the counting station until sufficient pressure is applied to the face of the container so that it conforms to the face of the photomultiplier detection device, and

axially shifting said photomultiplier detection device away from the container after completion of the count.

8. A method as claimed in claim 4 wherein said flexible containers include at least one face conforming in geometry to the surface of said photomultiplier detection device.

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