

Aug. 2, 1966

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H. LEVIN ET AL
RADIATION STERILIZATION DEVICE WITH
UNIFORMLY DISTRIBUTED BETA EMITTERS

3,264,473

3 Sheets-Sheet 1

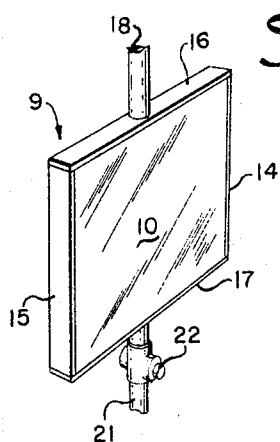


Fig 1

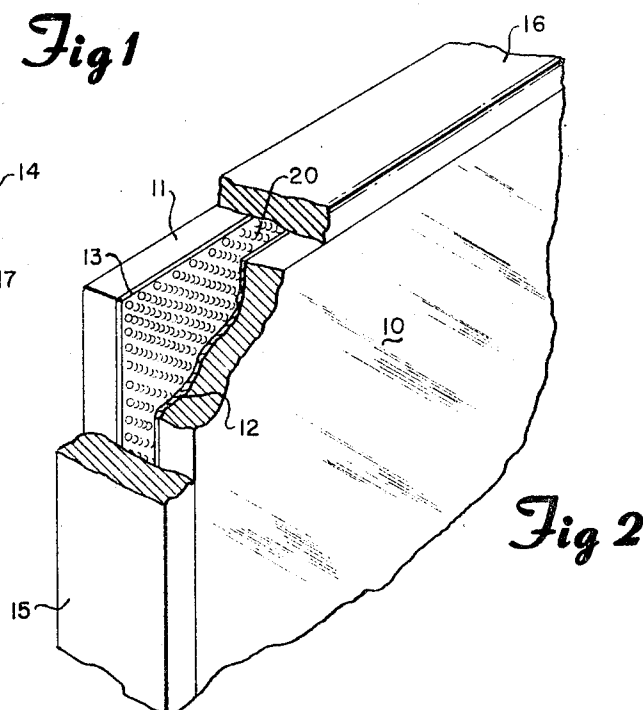


Fig 2

Fig 4

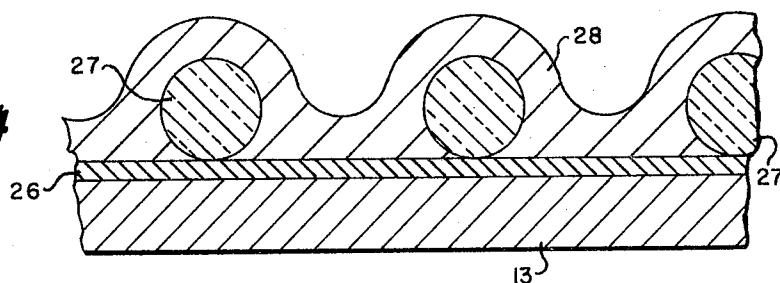
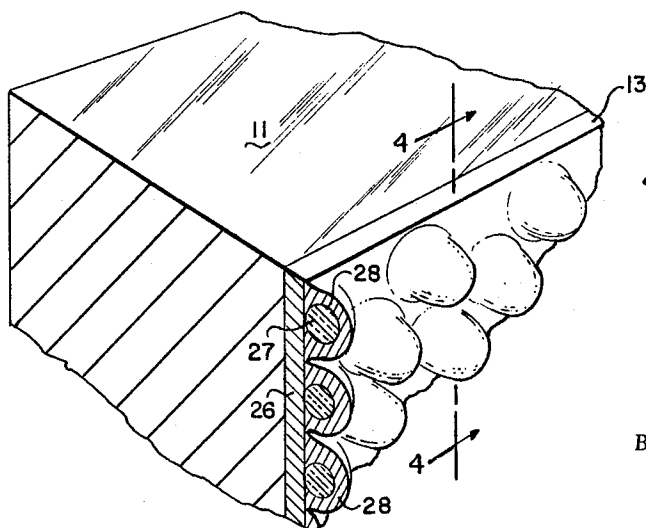


Fig 3

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3 Sheets-Sheet 2

Fig 5

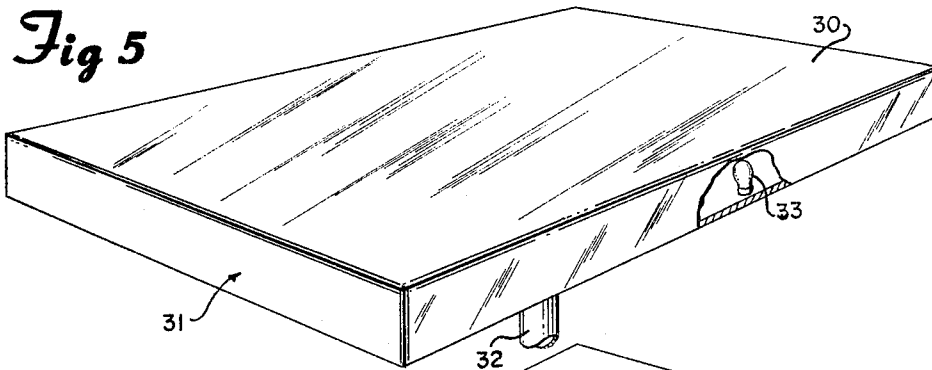


Fig 6

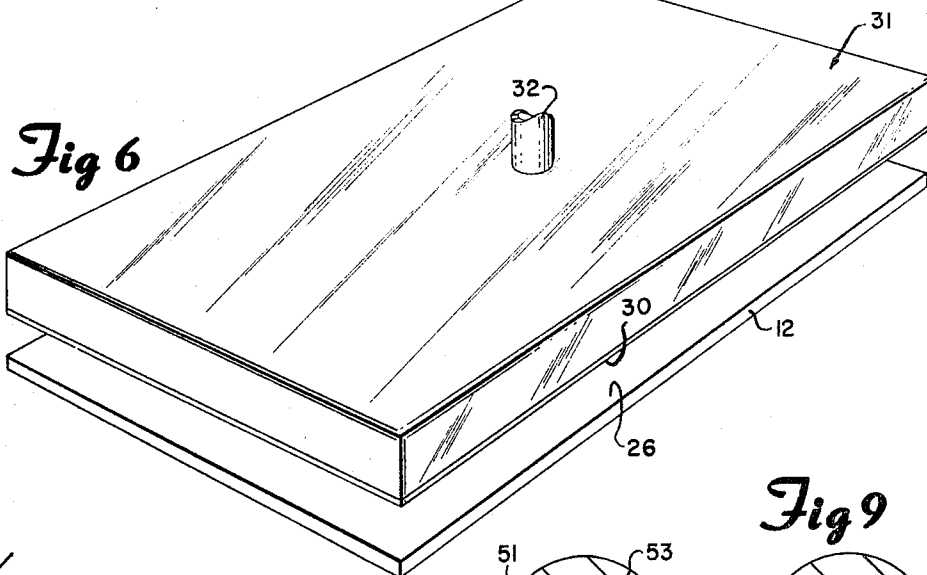


Fig 9

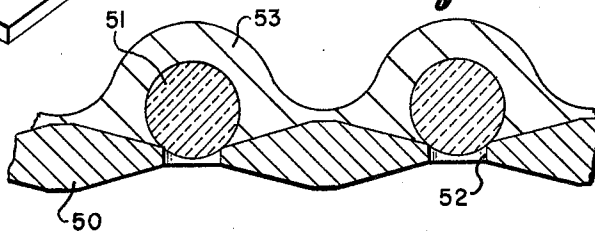
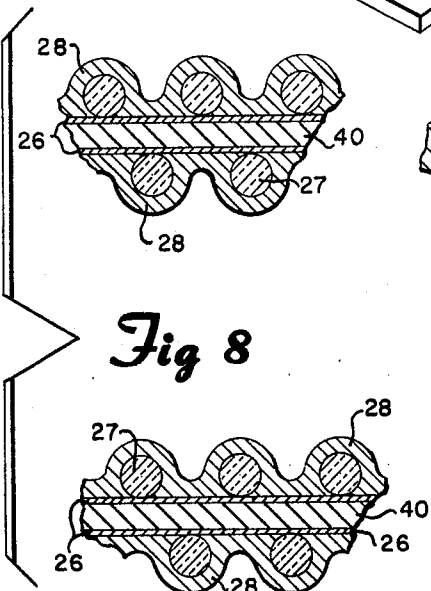


Fig 8



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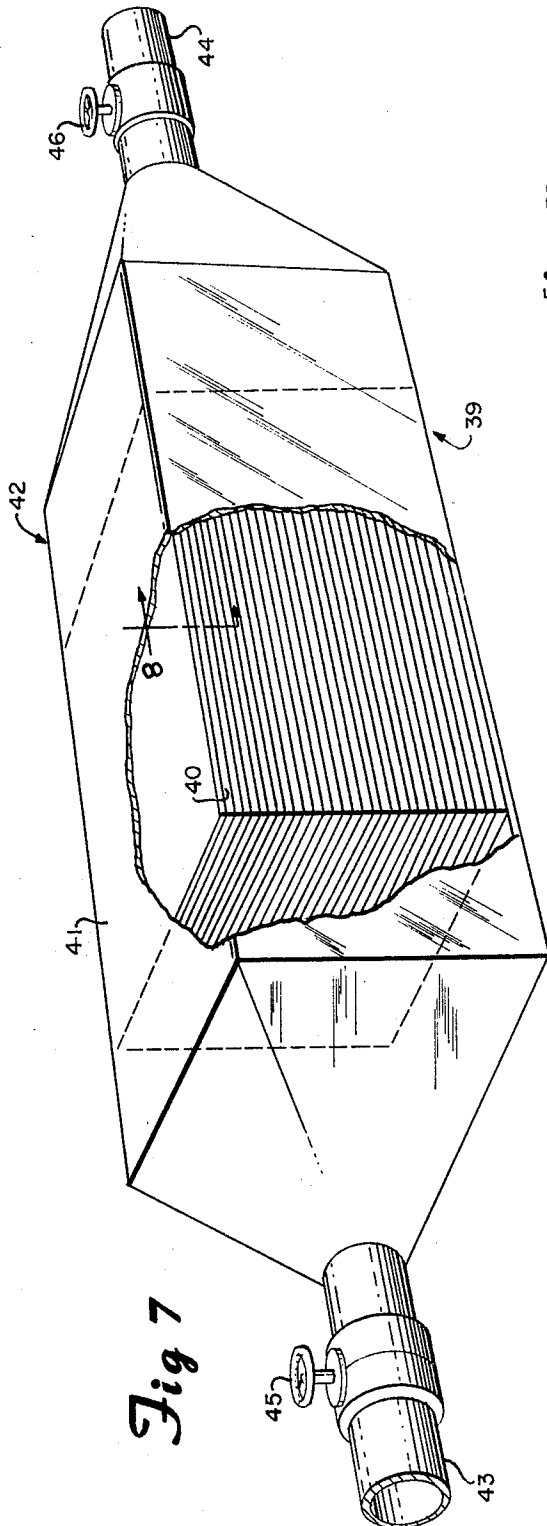


Fig 7

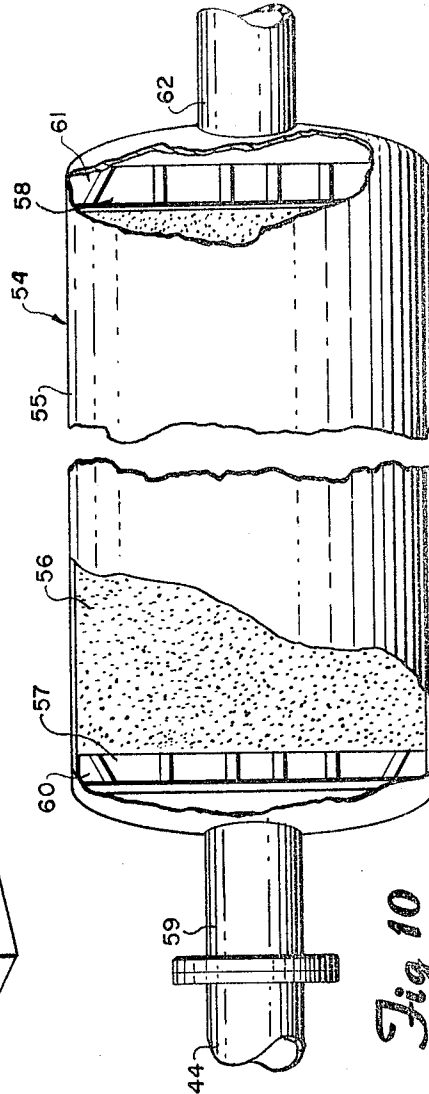


Fig 10

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RADIATION STERILIZATION DEVICE WITH UNIFORMLY DISTRIBUTED BETA EMITTERS

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13 Claims. (Cl. 250-44)

This invention relates to a radiation sterilization device, and more particularly to a sterilization device in which a beta emitter in the form of small coated ceramic particles, uniformly distributed over the surface of a plate, is utilized to uniformly emit beta radiation and thereby provide maximum efficiency of operation with a minimum of shielding.

Sterilization devices presently in use depend almost entirely upon the batch treatment with strong radioactive isotopes (such as strontium-90) which must be stored in a shielded compartment under lock and key and which must be handled in a special manner when removed from storage to treat the material to be sterilized. In the present invention, the even distribution of the radioactive ceramic particles permits the use of a low level beta emitter, such as promethium-147, whose radiation can be controlled in a very precise manner. The small ceramic beads treated with promethium-147 (and with promethium-148 eliminated) are presently commercially available. While such beads have been randomly mounted on plates, the present invention depends upon the uniform distribution of the beads with a spacing and size such that the radiation particles can be utilized with maximum efficiency. By utilizing the pure beta emitter promethium-147, the problem of shielding is minimized since the beta particles themselves are regarded as safe to the humans, in the absence of being ingested, and only weak X-rays are produced by the beta particles.

It is therefore an object of the present invention to provide a radiation sterilization device which utilizes very small particles of a radioactive isotope uniformly distributed on a plate to produce maximum efficiency in the use of the emitted particles.

Another object of the invention is to provide a sterilization device having a plate on which radioactive particles are distributed at an optimum distance from each other to provide a uniform, low level radiation field for the treated matter so that a maximum of the radiation energy is utilized in the sterilization process.

Another object of the invention is to provide a radiation sterilization device in which ceramic beads of promethium-147 are distributed on a mounting plate and the size and spacing of the beads are selected to provide minimum absorption between beads (which takes place excessively if the beads are too small) and to provide minimum self-attenuation (which takes place excessively when the beads are too large).

These and other objects of the invention not specifically set forth above will become readily apparent from the accompanying description of the drawings in which:

FIGURE 1 is a perspective view of a small sterilization device incorporating the present invention and capable of batch or continuous operation;

FIGURE 2 is an enlarged view of one corner of the device of FIGURE 1 which has been broken away;

FIGURE 3 is a greatly enlarged view of one of the radiation surfaces of the device of FIGURE 1, showing the coating films surrounding each of the beads;

FIGURE 4 is a cross-section taken along line 4-4 of FIGURE 3 showing the mounting plate and the manner in which the beads are mounted thereon;

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FIGURE 5 is a perspective view of the template and vacuum box utilized for evenly distributing the ceramic beads on the mounting plates;

FIGURE 6 is a view of the template of FIGURE 5 in inverted position and ready to place the evenly distributed beads on the tacky film deposited on the sterilization plate;

FIGURE 7 is a perspective view, partially in section, showing another form of the sterilization device incorporating the invention and having an increased flow capacity;

FIGURE 8 is a sectional view through two adjacent plates of FIGURE 7 showing the manner in which the beads are staggered on each plate;

FIGURE 9 is a modified form of mounting for the ceramic beads which permits effective radiation from the beads in both directions from the mounting plate; and

FIGURE 10 is an elevational view, partly in section, of an ion exchanger which can be connected with the discharge passage of FIGURE 7.

The embodiment of the invention chosen for illustration in FIGURES 1-3 comprises a device 9 comprising a rectangular-shaped container having stainless steel side members 10 and 11 which support mounting plates 12 and 13, respectively. The mounting plates have opposing faces which are coated with ceramic particles (beads) which have been treated with promethium-147. The side members and plates are held apart by side strips 14 and 15 and by top and bottom strips 16 and 17, all of which are secured in a suitable manner to the edges of the members 10 and 11. The plate 16 contains an opening for the inlet passage 18 which introduces a fluid to be treated into the space 20 between the plates. This fluid flows from the space 20 through the passage 21 which is controlled by a valve 22. All of the passages and fittings are constructed of stainless steel. Each of the plates 12 and 13 consist of stainless steel approximately 3 mils thick and each plate is covered with a coating 26 of porcelain or vitreous enamel which is approximately 1 mil thick. The ceramic beads 27 are evenly distributed on the coating 26 and then covered with another vitreous, porcelain or vapor deposited metal coating 28 which is approximately 2 mils thick. The coating 28 serves to retain the beads on the plate 11 and also provides protection against their leaching into the surrounding material in space 20. A device with the dimensions set forth above is shown in actual size in FIGURE 1 and has a 2-milliliter capacity which provides a highly useful analytical tool for laboratory use. The beads 27 have a mean diameter of about 150 microns (.150 mm.) and comprise a suitable ceramic material into which has been induced a quantity of promethium-147 to provide a radioactivity level for the beads of approximately 200 curies per gram. The very small beads have a very precise and uniform distribution of about 7,740 per square inch of surface of plates 12 and 13. With this distribution and size of beads, the device of FIGURE 1 has a total activity of approximately 40 curies. The depth of space 20 between the plates 12 and 13 has a maximum dimension of about 1.3 mm.

In operation, the specimen liquids, solutions or dispersions are admitted to the unit 9 through passage 18 with the valve 22 closed, by a hypodermic syringe or pipette. Following the prescribed exposure time, the specimen is then removed from the device for subsequent examination or utilization through passage 21 by opening valve 22. However, instead of batch treatment, the valve 22 can be set to provide a continuous flow of the specimen through the space 20 at a desired rate to effect the desired radiation absorption. The invention is not limited

in application of the treatment of liquids since various closure designs may be incorporated to permit the placement and radiation of solid materials in the space 20.

The uniform distribution of the beads is in a hexagonal pattern with a spacing of 0.0114 inch which results in the area density of 7,740 beads per square inch. With the hexagonal pattern, the distance between individual beads is the same in all directions so that each bead has an equal sphere of influence with each of the other beads. The radiation level of 40 curies is sufficient to cover the levels required in sterilization and death of microorganisms and for analytical and quantitative fundamental research.

In the batch-type irradiation of a liquid specimen in device 9 as previously described, a 2.5×10^8 rad dose level can be obtained in 4.27 hours and other dose levels are obtainable by simply varying the time of exposure. In general, the number of rads that will be produced in sample specimens is determined by the formula

$$C \times E \times 3.7 \times 10^{10} \times E_{\text{eff}} \times \Delta t$$

where C is the curies, E is the energy in m.e.v., E_{eff} is the efficiency of absorption, and Δt is the exposure time. It is therefore possible, knowing the value of E and C and taking an efficiency of approximately 30%, to determine the length of time required to produce a required number of rads in the sample material, one rad being defined as 100 ergs (of energy) absorbed in one gram of material. The isotope promethium-147 is a pure beta emitter, having a half-life of 2.6 years, a maximum energy level of 0.223 m.e.v., and an effective energy of 0.069 m.e.v. While the surfaces of plates 12 and 13 will be slightly dimpled because of the presence of the beads, the size of the beads on the plates is so small that a substantially uniform beta emission density will result outwardly from the plates.

When a water solution is to be introduced into the device 9, the separation of 1.3 mm. between the plates results in the absorption of practically all of the beta particles which are emitted outwardly from the beads. Beta particles of effective energy of .069 m.e.v. will penetrate .9 mm. of water and since the spacing between the plates is 1.3 mm., most of the particles entering space 20 from each of the plates 12 and 13 will be absorbed. As a general proposition, it is desirable to have a maximum amount of beta particles lost in the sample and to have a uniform density of particle absorption across the space 20. Also, the 30% efficiency factor takes into consideration that approximately 50% of the particles go back into plate 12 or 13 and another portion of the particles are absorbed by the coating 28. Thus, an efficiency factor in the range of 20-30% is considered to be acceptable. These efficiencies of promethium-147 betas and comparative efficiencies for other beta emitters are higher than obtainable in prior devices and results from the fact that each particle has an equal sphere of influence because it is equally spaced from all other beads. Also, the bead spacing is such that optimum distance is provided to obtain the maximum radiation from each bead with minimum absorption in adjacent beads, and the bead size is such as to provide minimum self-attenuation of the beta particles.

The beta emission from promethium-147 produces X-rays as it travels through the component structures of device 9 and these X-rays are weak in contrast to those produced by other beta emitters. The significance of the weak X-rays is that the shielding requirement is either absent or requires the presence only of a thin plate of plastic between the user and the source. Actually, less than 0.5% of the radiation energy escapes as X-rays from the device 9.

Several methods have been developed for the even distribution of the ceramic beads over the surface of the plates 12 and 13. Referring to FIGURES 5 and 6, a template 30 is attached to the open top of a rectangular prismatic vacuum box 31 in any suitable manner and

box 31 has an opening in its bottom to receive vacuum line 32. The template 30 is a sheet of thin-gauge sheet metal plate in which extremely small and very closely spaced holes are made by an electroetic process known as "electro-forming." When finished, the perforated plate appears as a very fine mesh screen with uniformly spaced holes. By use of this process, the holes in the metal plate can be as small as 5 microns in diameter and can be spaced closely enough to occupy over one-half of the plate's surface. Furthermore, the holes can be formed having a circular or other shape and in depth they can be made cylindrical, conical or other special shapes. For radiation device 9 of FIGURE 1, the template plate has circular holes 135 microns in diameter spaced in a regular hexagonal pattern 7,740 to the square inch.

The template assembly, including the plate 30 attached to the box 31, is then placed in an adapter retainer (not shown) in the form of a slightly larger open-top box with sidewalls to retain the beads and equipped with a small electrical vibrator. The beads of 150 micron mean diameter are placed upon the template 30 and a vacuum is pulled on the assembly while the adapter retainer is vibrated. Any template holes not filled by beads are detected by a light source 33 from within the vacuum box and these holes are filled by a manually operated syringe or piston-type applicator. After all holes are filled, the complete template assembly is removed from the retainer and inverted while the vacuum keeps all beads in place in the template holes. Meanwhile, a radiation plate 12 has been prepared for bead deposition by applying the 1-mil thick porcelain or vitreous enamel coating 26. The thickness of this deposition can be precisely controlled by known techniques, such as spraying through electrically charged screens. The coating 26 is dried in a dust-free environment to a tacky condition.

Thereafter, the inverted template (see FIGURE 6) is applied to coated plate 12 with sufficient pressure to cause the bead to stick to the coating 26. The vacuum is then released and the template removed, leaving the beads deposited in a regular pattern on plate 12. The radiation plate 12, with beads 27 attached to coating 26, is then placed on a rotating table and sprayed again with porcelain or vitreous enamel to uniformly cover the beads with a 2-mil thick coating 28. The radiation plate is then oven fired to a glassy finish. It is of course apparent that the beads can be applied to both sides of the radiation plate if such a structure is desired, by repetition of the process. After the oven firing of the plate, the plates are ready for assembly on side members 10 and 11 for use in radiation tests.

In general, the larger the size of the ceramic beads, the greater the emission and the fewer number of beads per square inch will be required. There is a range of bead sizes and beads per square inch which will produce a given radiation level most efficiently. If the beads are too small and too many, too much interference exists between beads and the beads will disperse the radiation into each other. On the other hand, if the beads are too large and too few, there can be too much self-attenuation in the beads themselves and the radiation field will not be as uniform as desired. In the device 9 of FIGURE 1, a range in bead size from 35 microns to 150 microns would produce satisfactory efficiency and for any given radiation level the beads per square inch would increase as the bead size is decreased. Further, the activity of the beads can range from 100 to 200 curies per gram.

While the embodiment of FIGURE 1 utilizes beads 27 treated with promethium-147, it is understood that beads treated with strontium-90 or other stronger beta emitter can also be utilized and fewer beads would then be required for the same radiation level across the space 20. It is also understood that the depth of space 20 can be varied with the type of material to be treated, since the penetration of the beta particles is a function of the

type of specimen being treated. Also, variation in space 20 will vary the percentage of emitted particles which are absorbed by a given material. The coating layer 28 prevents leaching of the radioactive material into the treated specimen. In the event it is desired to shield the device 9 under conditions where it will be manipulated over a long period of time, a plastic sheet of $\frac{1}{4}$ " to $\frac{1}{2}$ " thickness placed between the operator's body and the device will be sufficient to stop any X-rays emitted from device 9 as a result of beta particles striking the hard stainless steel casing.

The template process of producing the distribution of beads on the plates provides a high degree of reproducibility and tolerance control of radiation level. Although small individual variations occur among the beads, a high degree of uniformity of dose application is obtained because of the high surface density of the beads. For a small increment of volume of the irradiated sample, a uniform and accurately calculable dose can be achieved. The total radiation dosage can be accurately controlled by control of exposure time. By immersion of the radiation plates in a liquid phosphor or other scintillation medium (in conjunction with photo multiplier, amplifier, scalar instrumentation), the radiation dosage from each plate can be determined to a high degree of exactitude. The calibration of each plate can be carried out as a step in the manufacture of the device. The date and time of calibration can be stamped on the device, and the dosage rates (corrected for time-dependent radioactivity decay) can be precisely known for any time thereafter without further calibration. While less than .5% of the radiation energy (as X-rays) escapes from the device, this escape factor can be precisely calibrated during the manufacture of the device by measurement with various density specimen materials.

The device 9 is simple, small, lightweight, portable, and requires no external power for operation, has no moving parts, and can be operated in a zero gravity environment. The device is designed primarily as an analytical and quantitative tool for fundamental biomedical, biochemical, and biophysical researches. By virtue of the precise irradiation, it provides means for conducting experimentation (a) of controlled genetic changes in micro-life forms, seed, etc.; (b) of changes in biochemical compounds at specific dose levels; (c) of the effects of radiation dose levels on viruses, bacteria, yeast, molds, enzymes and other chemically and metabolically labile systems; and (d) of changes in physical materials, such as plastics, elastomers, etc.

Referring to FIGURES 7 and 8, there is shown another form 39 of the invention which utilizes a multiple of spaced mounting plates 40, each 3 mils thick, located within the rectangular portion 41 of stainless steel casing 42. Any suitable structure (not shown) can support the plates within the casing in spaced relationship. As shown in FIGURE 8, each plate 40 has the coating 26 of 1 mil thickness on each side for initially securing the beads 27 and the coating 28 of 2 mils thickness surrounds each of the beads on each side of a plate. The beads 27 on each side have a 150-micron mean diameter and 7,740 beads per square inch are arranged in a hexagonal pattern so that the structure on each side of the individual plates is the same as that shown on the single plate side in FIGURE 4. The beads are staggered on opposite sides of each plate for minimum absorption by the beads themselves and to provide minimum obstruction to passage of radiation from the beads on one side of the plate to the medium to be irradiated on the opposite side of the plate. The plates are uniformly stacked in sufficient quantity and with the requisite spacing between each plate to provide effective deactivation and sterilization of the passing fluid at the design flow rate. The fluid to be treated enters the passage 43 and is uniformly distributed between the plates by virtue of the equalization of pressure across the plates at the very low flow rate. The

case 42 is maintained full of treated solution, while over-flow through tube 44 is continuous at the design flow rate. Valves 45 and 46 serve as means for adjustment of flow. There may be up to 100 or more plates 40 within the stack, and the flowing liquid, to be treated by the promethium-147 beads of approximately 200 curies per gram activity, passes between the plates 40 and encounters the emitted radiation from the beads and thereby undergoes deactivation and/or sterilization. To permit effective use of the sterilization between the adjacent plates, the spacing between adjacent plates is about 1.2 mm. Because of the extremely small diameter of the beads and the very close spacing of them on the plate 40, a highly uniform distribution of the relatively weak beta radiation is directed into the flowing fluid. For this reason, and for reason of low flow rate, no "channeling" occurs which might prevent some portions of the fluid from receiving equal radiation treatment.

It will be obvious that each side of the plates 40 of the device of FIGURE 7 can be prepared by the same method as the plates 12 and 13 of the prior embodiment. If desired, each plate 40 can be replaced by a plate 50, which is shown in FIGURE 9, and the beads 51 on plate 50 are permitted to radiate from both sides of the plate. Before coating with beads, the plate 50 is the same as template 30 shown in FIGURE 5. In fabrication, once the beads 51 fill all the template holes 52 in the plate 50, they are held therein by the vacuum and the excess beads are removed, and while the vacuum holds the beads in place, the template is then sprayed with a suitable material to provide a coating 53 for retaining the beads on the plate. It is noted that this configuration permits a more effective release of radiation from the beads where it is desired to irradiate the fluid on both sides of the supporting plate. Advantage is taken of the fact that the electro-forming process can form holes in the plate with chamfered edges as shown in FIGURE 9. This radiation plate configuration allows a greater amount of radiation from the bead to reach the flowing fluid on the wall sides since the metal surrounding the holes is extremely thin.

The device 39 can be designed to have a flow rate of 2 grams per minute but, of course, a wide range of flow rates may be achieved by applying suitable sealing factors to the device. The device is particularly useful for processing human waste for possible water recovery, for biological fuel cell utilization and for other applications that demand enzyme deactivation and/or sterilization. The human waste material is first collected and processed prior to being admitted into the line 43 under suitable pressure and as a homogenous fluid mixture or solution.

In the event that potable water is required from device 39, the component ion exchanger 54 shown in FIGURE 10 can be attached to the outlet tube 44 of FIGURE 7 for further processing of the fluid. The exchanger 54 has a casing 55 containing a strong ion exchanger resin 56, such as a sodium resinate. Promethium-147, which might leach from the beads in the sterilized device 39 and dissolve in the treated solution, is captured by the resin which is in the form of beads of approximately 40-mesh size held tightly as a packed bed between screens 57 and 58. The inlet passage 59, connected to passage 44, introduces the fluid to a filter-diffuser plate 60 and a diffuser plate 61 is downstream of screen 58. The discharge of potable water is from passage 62. While promethium-147 is a toxic if ingested by humans (the maximum permissible concentration in water being 2×10^{-3} microcuries per cubic centimeter for a 168-hour week), it is regarded as safe for external use when properly confined.

In both forms of the invention, the extremely small diameter of the beads and the very close spacing of them on the plate provide a highly uniform distribution of the relatively weak beta radiation which is directed into the material between the plates. Since the spacing of the particles is uniform, each bead has the same sphere of in-

fluence and substantially uniform radiation activity is produced between the plates. By coating the individual particles as they are placed on the plates, leaching of the toxic radioactive material into the solution is prevented. At the same time, the coating served to hold the particles or beads on the plate while permitting passage of a large percentage of the beta energy into the solution. The weak beta emission from promethium-147 produces X-rays that are very weak so that little or no shielding is required for safety purposes. Generally, the invention provides for the maximum utilization of the beta particles omitted from a minimum amount of material, thereby utilizing a minimum of beta radiation to accomplish a desired radiation level. The particular dimensions, configuration and capacities referred to herein are solely for purposes of illustration and it is understood that these quantities can be varied to meet a particular requirement, whether for a batch or continuous flow process. For example, while a 150 micron mean diameter particles is disclosed for the two embodiments, the bead size can be varied over a range of sizes. By selecting the size and spacing of the beads to obtain maximum efficiency and utilization of the beta emission, the shielding requirement is eliminated or greatly reduced. When other beta emitters such as strontium-90, are utilized, the bead size and spacing can be optimized in a like manner as described in connection with promethium-147 to provide a spacing, configuration, bead surface density, etc. which will provide maximum efficiency in the use of the radiation. While the same general considerations can be applied to other beta emitters, it is understood that the energies and penetrations are different for each beta emitter. Various other modifications are contemplated by those skilled in the art without departing from the spirit and scope of the invention as hereinafter defined by the appended claims.

What is claimed is:

1. A radiation sterilization device comprising:
a mounting plate;
means associated with said plate for confining a material adjacent said plate for treatment;
a plurality of particles located on said plate in a hexagonal pattern for uniform distribution of said particles, said particles being treated with a radioactive isotope so that each particles is a radioactive emitter, and;
a coating over said individual particles for resisting leaching of said particles into said treated material while permitting radiated energy to enter into said material.
2. A radiation sterilization device as defined in claim 1, wherein said particles are ceramic particles treated with isotope promethium-147 to emit beta radiation.
3. A radiation sterilization device as defined in claim 2 wherein said ceramic particles are treated to emit beta radiation with an activity in the range of 100 to 200 curies per gram.
4. A radiation sterilization device as defined in claim 2 wherein said ceramic particles have a uniform size in the range of 35 to 150 microns mean diameter.
5. A radiation sterilization device as defined in claim 2 wherein said particles have an activity of 200 curies per gram and a size of 150 microns, the particles being uniformly spaced apart 0.0114 inch to give a particle density of 7,740 particles per square inch.
6. A radiation sterilization device as defined in claim

1, wherein said coating comprises an enamel coating of approximately 3 mils thickness.

7. A radiation sterilization device as defined in claim 1, wherein said particles are deposited on an enamel coating on said mounting plate of approximately 1 mil thickness.

8. A radiation sterilization device comprising:

a plurality of particle mounting plates;
means for confining a material adjacent said plates for radiation treatment;

a plurality of particles located on the surface of each plate in a hexagonal pattern for uniform distribution of said particles, said particles being treated with a radioactive isotope so that each particle is a radioactive emitter, and

a coating over said individual particles for resisting leaching of said particles into said treated material while permitting radiated energy to enter into said material.

9. A radiation sterilization device as defined in claim 8 wherein said plurality of plates comprises a pair of mounting plates with opposed radiating surfaces, said confining means comprising panel means extending around the edges of said plates.

10. A radiation sterilization device as defined in claim 7 wherein said particles have approximately a 150-micron mean diameter and the spacing between said pair of plates is approximately 1.2 millimeters.

11. A radiation sterilization device as defined in claim 8 wherein said mounting plates are arranged in a stack with the radiating surfaces spaced apart and parallel to one another, said confining means comprising a casing surrounding said stack.

12. A radiation sterilization device as defined in claim 8 wherein said confining means contains opposed openings for receiving inlet and outlet passages for said material, valve means in one of said passages for controlling the time of treatment of said material, and an ion exchanger connected with said outlet passage to capture any particle material leached into the treated material.

13. A radiation sterilization device comprising:

a mounting plate positioned adjacent a material to be treated;

a plurality of particles uniformly distributed on said plate, said particles being treated with a radioactive isotope so that each particle is a radioactive emitter; and

a coating over said individual particles for resisting leaching of said particles into said treated material while permitting radiated energy to enter into said material.

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