



US 20090274258A1

(19) **United States**

(12) **Patent Application Publication**  
**Holden et al.**

(10) **Pub. No.: US 2009/0274258 A1**

(43) **Pub. Date: Nov. 5, 2009**

(54) **COMPOUND ISOTOPE TARGET ASSEMBLY FOR PRODUCTION OF MEDICAL AND COMMERCIAL ISOTOPES BY MEANS OF SPECTRUM SHAPING ALLOYS**

filed on Apr. 14, 2006, provisional application No. 60/792,066, filed on Apr. 14, 2006, provisional application No. 60/805,541, filed on Jun. 22, 2006.

(76) Inventors: **Charles S. Holden**, San Francisco, CA (US); **Robert E. Schenter**, Portland, OR (US)

Correspondence Address:  
**STAINBROOK & STAINBROOK, LLP**  
**412 AVIATION BOULEVARD, SUITE H**  
**SANTA ROSA, CA 95403 (US)**

(21) Appl. No.: **12/296,725**

(22) PCT Filed: **Apr. 13, 2007**

(86) PCT No.: **PCT/US07/66666**

§ 371 (c)(1),  
(2), (4) Date: **Oct. 10, 2008**

**Related U.S. Application Data**

(60) Provisional application No. 60/792,067, filed on Apr. 14, 2006, provisional application No. 60/792,065,

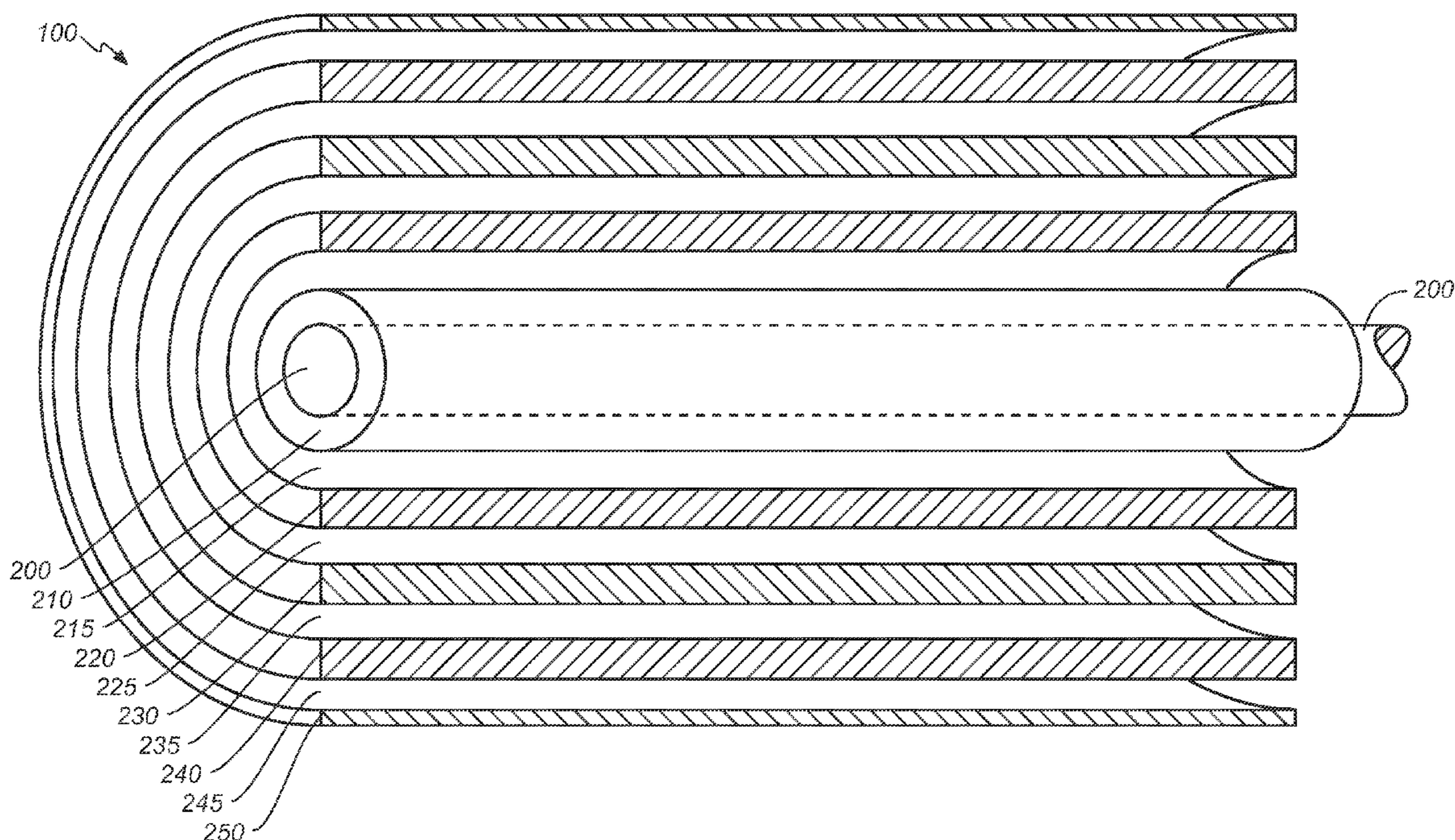
**Publication Classification**

(51) **Int. Cl.**  
**G21G 1/00** (2006.01)

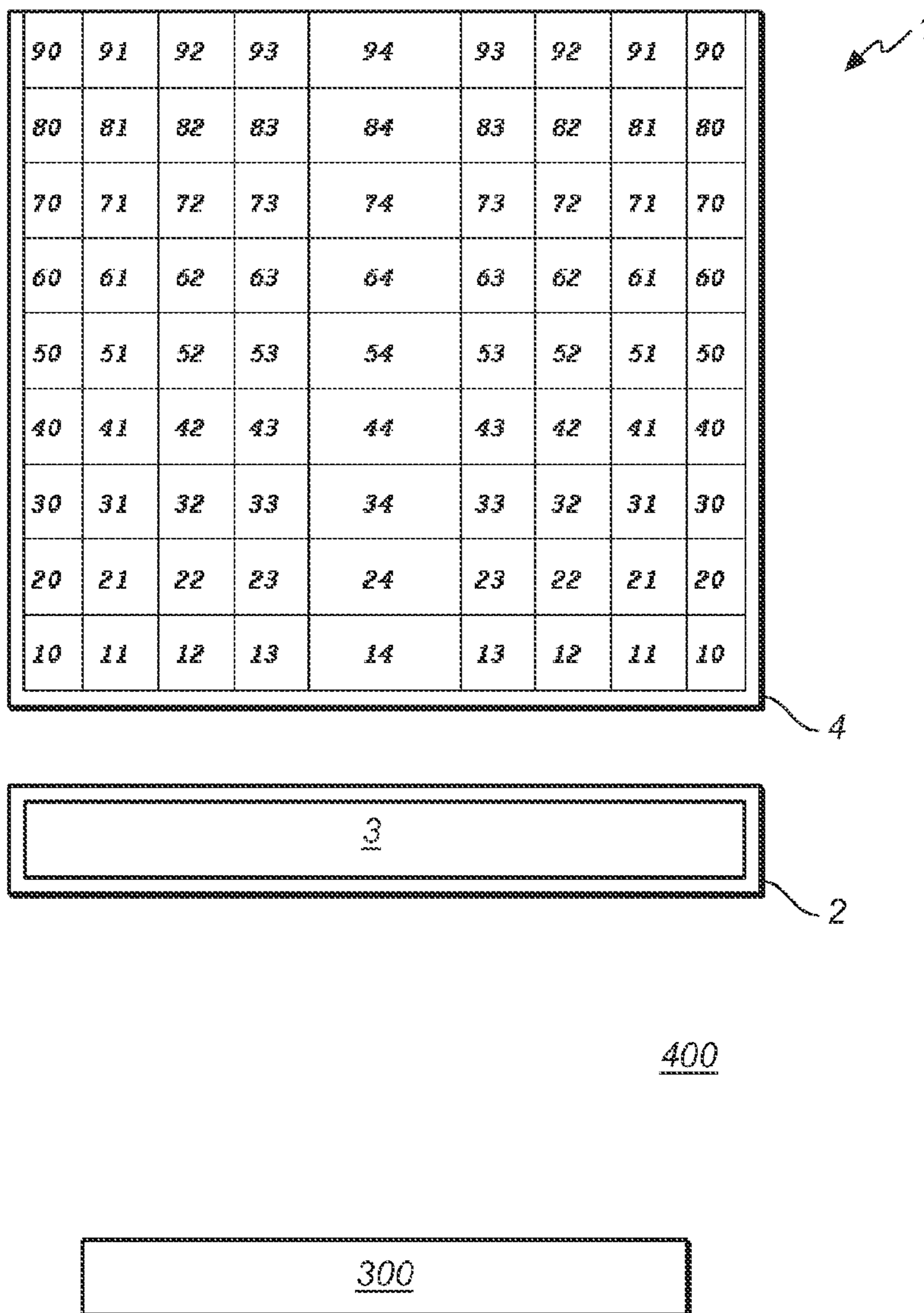
(52) **U.S. Cl.** ..... **376/190**

(57) **ABSTRACT**

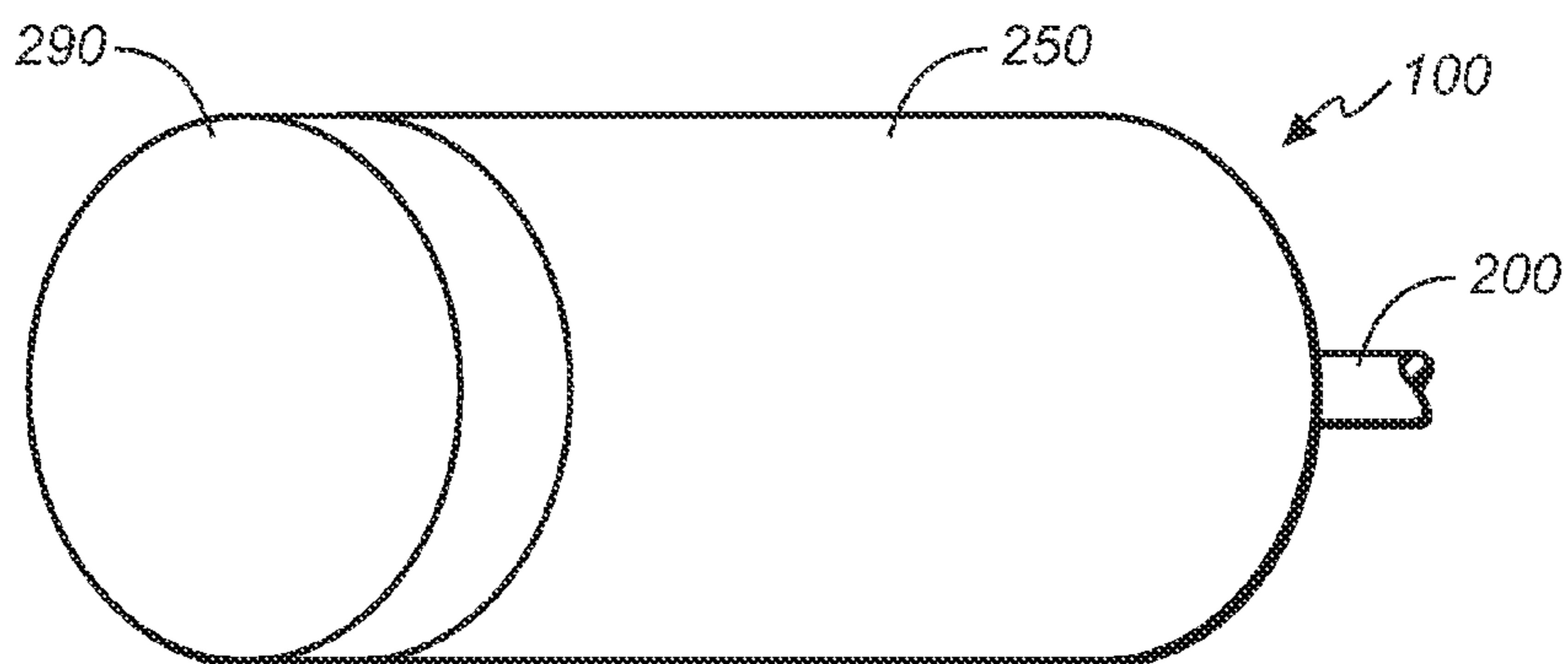
A neutron generator and method of using the same to produce commercial and medical isotopes. Isotopes are transmuted using fission spectrum neutrons produced in neutron multiplier alloys proximate a target isotope. The produced neutron fission spectrum is tailored or shaped by alloys disposed between the multiplier alloys and the target isotope. The tailoring alloys selectively convert the neutron fission spectrum to a nearly coherent distribution of selected high energy, fast, epithermal, or thermal neutrons. The tailoring alloys are engineered to slow the fission spectrum neutrons to the resonance energies of the selected target isotopes through scattering or hydriding of selected components of high temperature aluminum alloys that optimize neutron capture in engineered target alloys.



**FIG. 1**



**FIG. 2**



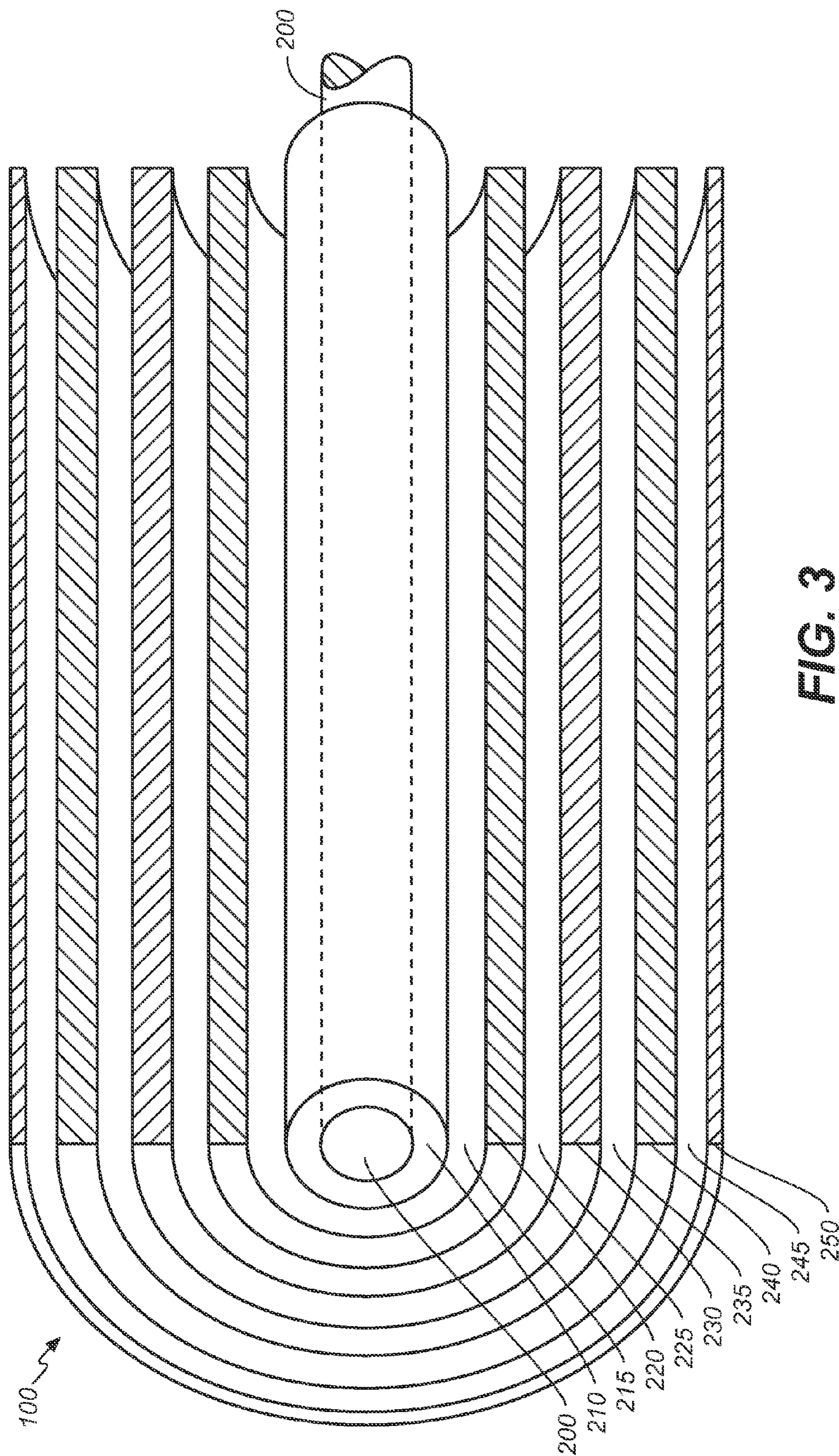
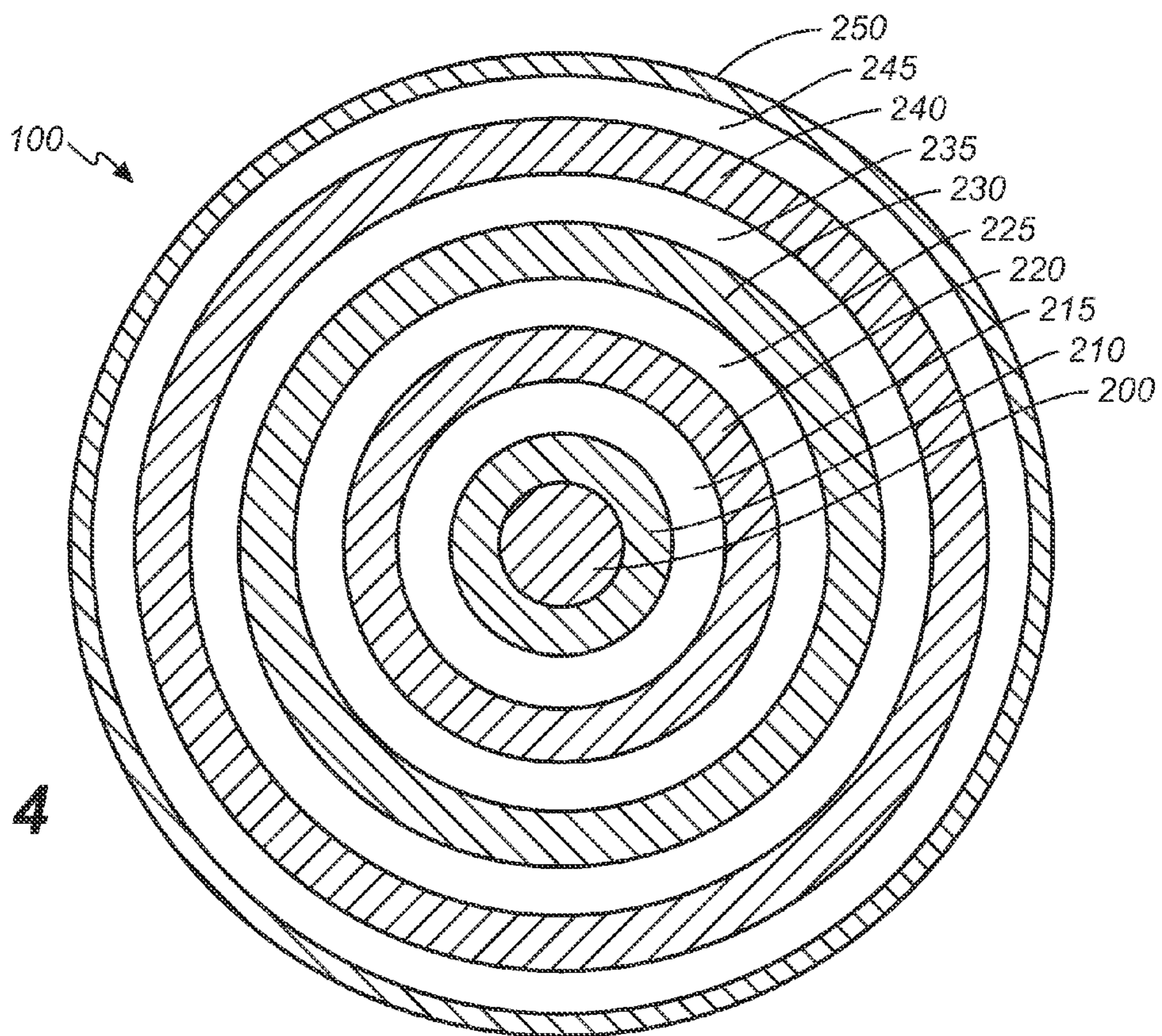
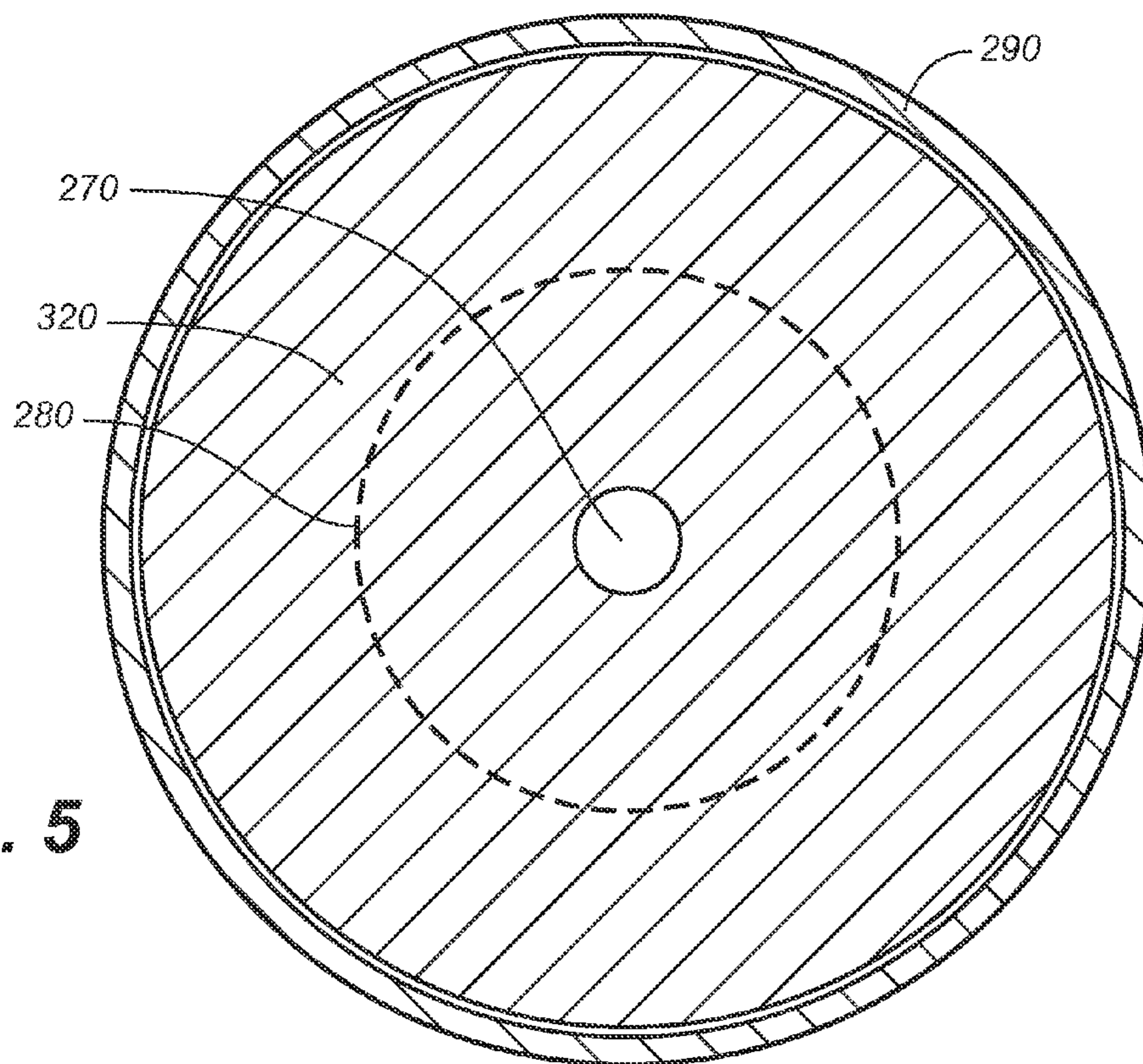


FIG. 3



**FIG. 4**



**FIG. 5**

**COMPOUND ISOTOPE TARGET ASSEMBLY  
FOR PRODUCTION OF MEDICAL AND  
COMMERCIAL ISOTOPES BY MEANS OF  
SPECTRUM SHAPING ALLOYS**

BACKGROUND OF THE INVENTION

**[0001]** 1. Technical Field

**[0002]** The present invention relates generally to means for producing commercial and medical isotopes, and more particularly to a method and apparatus for transmuting isotopes using fission spectrum neutrons produced in novel neutron multiplier alloys adjacent to the target precursor isotope, shaping or tailoring the produced neutron fission spectrum using novel alloys located between the multiplier and the target so as to selectively convert the neutron fission spectrum to a nearly coherent distribution of selected high energy, fast or epithermal or thermal neutrons and to slow down the fission spectrum neutrons to the resonance energies of the selected target isotopes.

**[0003]** 2. Background Art

**[0004]** It is well known that neutrons produce valuable isotopes for use in scientific research, manufacturing and medicine. Radioactive isotopes are employed in scientific research in fields as different as hydrology and life sciences. Isotopes are used in business and commerce in many manufacturing process and in the production of oil and gas. The most valuable and some of the most difficult to make isotopes are used in the diagnosis and treatment of human diseases and disorders.

**[0005]** It is also well known that fission in nuclear reactors produces fission products that are a component of nuclear waste. Accordingly, it would be desirable to provide means to transmute fission products that remain radioactive for a long time into shorter lived isotopes or stable isotopes.

**[0006]** Isotopes are currently produced by electron beams, ion beams, in cyclotrons, and in nuclear reactors. Some isotopes may be produced by any of the four general methods. Others by one method alone. The science of isotope production involves adding or subtracting the requisite number of nucleons (protons or neutrons) to or from a target parent isotope to produce the desired end product in significant quantities and with appropriate purity.

**[0007]** The transmutation reactions that change the number of protons or neutrons in the isotopes include photonuclear reactions wherein energetic gamma photons eject neutrons from nuclei. Also high energy neutrons are able to displace or eject protons from nuclei. Transmutation reactions caused by accelerated ions such as protons, deuterons, helium nuclei and other ions collide with the nuclei of the target material to change the target material from one isotope to another. These reactions generally do not yield the quantity of desired product material as efficiently as reactions involving successive or singular neutron capture in the nuclear reactor. Additionally, these reactions are not as efficient in the treatment of fission products to transmute them to shorter lived isotopes or common stable isotopes.

**[0008]** Fission reactors provide the most convenient environment for the production of medical isotopes, as this is where the requisite number of energetic neutrons is most easily produced, where they can be confined, and where adequate shielding against gamma radiation presently exists. Fission reactors also provide a suitable location for the transmutation of fission products produced in spent nuclear fuel.

**[0009]** Fission reactors in the United States and Canada are generally used for and available for irradiation of target materials for medical isotope production operate primarily in the thermal neutron spectrum, limiting the isotopes products efficiently produced to those with precursor capture neutrons in the thermal energy spectrum. For isotopes needing a more energetic neutron spectrum to efficiently produce the desired isotope, the thermal fission spectrum has shortcomings. Accordingly, many potentially useful isotopes exist in scant quantity insufficient for properly managed medical clinical trials. Higher energy charged particles are available in cyclotrons and accelerators but the high cost of beam time limits production to the isotopes in the highest demand since only charged particles can be accelerated. Therefore, a real need exists for improved novel methods to add fission spectrum, fast spectrum or epithermal neutrons successively or singly to selected target nuclei in existing reactors: test or experimental or university reactors to bring isotopes to the market in quantities large enough to meet scientific, medical and commercial demand.

**[0010]** The prior art that deals with methods to introduce isotopes into the patient for the treatment of disease reveal the value and importance of holmium-166, among other isotopes (see U.S. Pat. No. 6,767,531, to Fritzberg et al). The '531 patent teaches the benefits of numerous other isotopes that are discussed in medical literature cited as being efficacious for the diagnosis or treatment of diseases, and it provides detailed means and methods of the therapeutic application of a multitude of medical isotopes including holmium-166 into the persons afflicted with various forms of cancer and other diseases. The '531 patent does not disclose how to make holmium-166 and reveals no method for producing any medical isotope.

**[0011]** The foregoing prior art reflects the current state of the art of which the present inventors are aware. Reference to, and discussion of, this art is intended to aid in discharging Applicants' acknowledged duties of candor in disclosing information that may be relevant to the examination of claims to the present invention. However, it is respectfully submitted that none of the above-indicated patents disclose, teach, suggest, show, or otherwise render obvious, either singly or when considered in combination, the invention described and claimed herein.

DISCLOSURE OF INVENTION

**[0012]** The present invention is an apparatus and a method of using the same to produce a large quantity of specific activity radioisotopes in a single neutron capture mode or sequentially in a multiple capture mode within selected target isotope materials. The invention, in a first aspect, is a very compact "plug-in" type compound isotope target assembly ("capsule") that locally boosts the neutron energy spectrum and the neutron flux to provide more energetic neutrons proximate to the target isotope. This makes higher activity material as the desired end-product.

**[0013]** The first component of each of the preferred embodiments of the compound target assembly is a neutron multiplier alloy, the second is a spectrum shaping or tailoring alloy, and the third is the target precursor isotope to be transmuted. The multiplier supplies a rich flux of fission spectrum neutrons. These are slowed down in the spectrum shaping alloy to promote probabilities of capture in the precursor isotope or isotopes constituting the target.

**[0014]** The inventive compound target assembly preferably includes two volumes of neutron multiplier material, preferably disposed in an exterior tube and interior rod, and further includes successive rows or plates of differing material layers, which in the preferred embodiments are neutron multipliers, neutron spectrum shaping alloys, and fission product containers isolating the precursor isotope from fission product contamination by sandwiching it between power rods or plates.

**[0015]** The novel compound isotope target assembly employs fissile material in a set of inventive alloys or inert metal matrixes that seals the fission products in the multiplier alloys and allows the energetic fission spectrum neutrons to escape from this matrix without significant energy loss. Those neutrons are delivered through a spectrum shaping alloy placed on the inner and outer sides of the target isotope material.

**[0016]** The target material comprises a selected precursor isotope for each of the desired isotopes to be produced as well as a spectrum shaping alloy material so that the precursor material to be transmuted captures neutrons efficiently in the selected resonance energy regions. This maximizes the production of a desired isotope in the heightened flux environment. The spectrum shaping material can be a hydrided or deuterated alloy, an oxide strengthened alloy, or an alloy containing dispersed carbides or nitrides other neutron scattering material that shapes or tailors the spectrum appropriately. As will be readily appreciated, each spectrum shaping alloy is specifically formulated to shape or tailor the spectrum to maximize production of desired the medical isotope.

**[0017]** A scattering alloy of particular utility, and thus employed in a preferred embodiment, is one composed of very fine particles of boron-11 carbide (B<sub>4</sub>C) dispersed in aluminum silumen or vanadium aluminum zirconium alloys. This scattering alloy functions as a moderator slows neutrons in a controlled manner to reach the reduced energy needed for effective capture in the target material in the epithermal, fast, or (when provided without shielding or an intervening shaping alloy) in the high energy spectrum. Scattering materials can also be nickel, iron, vanadium, scandium, zirconium and other mid-Z metals.

**[0018]** The neutron multiplier uses thermal neutrons to fission uranium-233, uranium-235, plutonium-239, or a selected combination thereof with americium and curium if needed. The selected fissile material is dispersed in an inert metal matrix alloy that in some preferred embodiments also contains lead or beryllium to increase the local population of neutrons. The fissile material is in ceramic or metallic form. The neutron multiplier alloy consists of aluminum and zirconium and, if needed, refractory metals such as tantalum or depleted molybdenum or tungsten, as well as engineered quantities of beryllium and/or lead, bismuth, thorium or uranium. Thermal spectrum neutrons cause the fissile material to fission at a controlled rate. The fissile material is located in its highest concentration around the central axis of the power rod in the center and the exterior side of the power tube surrounding the array. For embodiments used out side of the reactor, an electron beam of 10 milliamps with electrons accelerated to 16 to 28 MeV interact with the electron target material in the neutron multiplier, or "converter." The electron target material in the converter is preferably an optimized alloy of tantalum, tungsten and/or rhenium to produce penetrating and energetic gamma photons. These photons interact with high-z nuclei to eject and produce neutrons. These neutrons are

moderated by spectrum shaping alloys to produce thermal neutrons. The thermal neutrons thus become available to the fissile material present, in optimized quantities. Produced neutrons and or fission spectrum neutrons pass through the second alloy, namely, the spectrum shaper, and enter the target at with a spectrum optimized for isotope production. The second alloy is therefore engineered for each target material. It consists of aluminum and zirconium for most applications where neutron capture is the desired object with dispersed hydrides, carbides or nitrides. The spectrum shaping alloy contains engineered amounts of scattering particles dispersed homogeneously in the alloy, examples of which are discussed below. The scattering materials are generally low-Z ceramics such as boron-11 tetra carbide (wherein the boron must be boron-11) or, lithium-7 boride (with boron as boron-11), lithium-7 nitride, boron nitride (with boron as boron-11eleven). These are low-Z materials with low neutron capture cross sections, which can be combined with hydrides of zirconium, titanium, vanadium, lithium to provide spectrum softening effects to optimize the spectrum for neutron capture in the selected target isotope.

**[0019]** The present invention can boost the comparatively high flux of existing nuclear reactors to enhance the higher and more energetic neutrons and higher flux to effect the desired transmutations when the capsule is placed into a reactor. In the preferred embodiments an electron beam interacts with a converter material to produce gamma photons which cause neutrons to be ejected from the nuclei of high-z isotopes such as lead, bismuth, thorium, or uranium. As noted, additional fission spectrum neutrons are generated in novel neutron multiplying alloys that use fissile materials in an inert metal matrix (and are similar to nuclear fuels) to produce a high flux of fission spectrum neutrons for many applications.

**[0020]** The method of producing the isotopes disclosed and claimed herein focuses on the production of medical isotopes whose precursor materials have large resonance integrals for capture of neutrons occurring in the epithermal energy region. However, the inventive method and apparatus are also appropriate for isotope production in targets that have large resonance capture in the fast spectrum 10 keV to 1 MeV. This includes: molybdenum-98, which yields molybdenum-99, which decays to technetium-99m. For epithermal capture, the following isotopes may be produced: holmium-165, that yields holmium-166; yttrium-89 that yields yttrium-90; ytterbium-176 that yields ytterbium-177 that decays to lutetium-177; and tungsten-186 that upon two neutron captures becomes tungsten-188 that decays to rhenium-188. Other medical isotope products include carrier free strontium-89, radium-223 and high purity iodine-131. Copper-64 and copper-67 are produced from zinc-64 and zinc-67 by the n,p reaction where high energy neutrons eject protons in target nuclei of the two zinc isotopes.

**[0021]** Note: As used herein, the thermal spectrum is defined as  $\frac{1}{1000}$  electron volts to  $\frac{5}{10}$  electron volts, the epithermal spectrum is  $\frac{5}{10}$  electron volts to 5000 electron volts, the fast as five thousand electron volts (5 keV) to one million electron volts (1 MeV) and the high energy spectrum as over one million electron volts (1 MeV) to twenty million electron volts (20 MeV).

**[0022]** There has, thus, been broadly outlined the more important features of the invention in order that the detailed description that follows may be better understood, and in order that the present contribution to the art may be better

appreciated. Additional objects, advantages and novel features of the invention will be set forth in part in the description as follows, and in part will become apparent to those skilled in the art upon examination of the following. Furthermore, such objects, advantages and features may be learned by practice of the invention, or may be realized and attained by means of the instrumentalities and combinations particularly pointed out in the appended claims.

[0023] Still other objects and advantages of the present invention will become readily apparent to those skilled in this art from the following detailed description, which shows and describes only the preferred embodiments of the invention, simply by way of illustration of the best mode now contemplated of carrying out the invention. As will be realized, the invention is capable of modification in various obvious respects without departing from the invention. Accordingly, the drawings and description of the preferred embodiments are to be regarded as illustrative in nature, and not as restrictive.

#### BRIEF DESCRIPTION OF THE DRAWINGS

[0024] FIG. 1 is a schematic view showing a first preferred embodiment (i.e., a two chamber design) of the neutron generator/isotope production apparatus of the present invention;

[0025] FIG. 2 is a perspective view of the compound target assembly (or "capsule") employed in a second preferred embodiment of the neutron generator apparatus of the present invention;

[0026] FIG. 3 is top cross-sectional perspective view thereof,

[0027] FIG. 4 is a cross-sectional end view in elevation of the target assembly of FIGS. 2 and 3; and

[0028] FIG. 5 is a cross-sectional end view in elevation of the central neutron generator cavity showing structure where the electron accelerator tube is attached to an electronics package and further showing the gamma generating alloy forming the cup and the selected target material for the gamma, n reaction.

#### BEST MODE FOR CARRYING OUT THE INVENTION

[0029] Referring to FIGS. 1 through 5, wherein like reference numerals refer to like components in the various views, there is illustrated therein a new and improved compound target assembly for the production of medical isotopes and commercial isotopes.

[0030] The drawings show the inventive compound target assembly. The structural elements of the assembly are fabricated from aluminum, zirconium, molybdenum, tungsten, vanadium, iron, tantalum or nickel in various combinations depending upon the desired product or application. These disclosed alloys "admit and conduct" thermal neutrons because the proportion of aluminum is high; other metals are added depending on the need for hydrogen or deuterium to thermalize neutrons, or the need to scatter the neutrons, or to raise the fission rate so that the assembly may function with an enhance neutron flux. Refractory metals are added to the alloy for safer service in conditions of elevated temperature.

[0031] The use of oxides for the fertile and fissile actinides are a preferred embodiment because no coating is needed and two can be placed together with thorium oxide as the fertile

material to promote negative reactivity as temperature becomes elevated and to provide the matrix to store the fission products.

[0032] Referring first to FIG. 1, there is shown in schematic form a first preferred embodiment of the inventive compound target assembly for a neutron generating and isotope production apparatus. This view shows that in an exemplary embodiment 1, an electron beam source 300 directs an electron beam to a first chamber 2, which is a converter cell, preferably comprising a hollow rectangular 2 mm tungsten alloy channel having an interior void 3 through which cooling water or other working fluid such as a liquid metal may be continually circulated to export heat and enable long term operation of the system. The electron beam source is separated from the converter cell by dry air 400, which surrounds the two chambers.

[0033] In the first preferred embodiment the system next includes a second chamber 4, which contains one or more target isotopes, and the volume of target isotope may be demised into a plurality of cells 10-13, 20-23, 30-33, 40-43, 50-53, 60-63, 70-73, 80-83, and 90-93, which receive various amounts of exposure to gamma rays at various energies. The energy dependent photon fluence may be calculated for each zone and optimized for the application selected.

[0034] FIGS. 2-5 show a second preferred embodiment of the inventive compound target assembly of the present invention, showing the compound isotope target assembly 100 comprising a central power rod 200 composed of neutron multiplying alloy, which is enclosed in a spectrum shaping alloy cup 210. The cup may be cooled by heavy water pumped through channels 215, exterior to the cup, as shown or by other coolants such as liquid metals, sodium or lithium, or liquid metal eutectics such as lead bismuth eutectic (LBE).

[0035] Axially disposed around the cup (and preferably spaced apart from the cup) is a tube, ring, or foil layer of precursor isotope 220, followed by an outer ring of spectrum shaping alloy 230, and an outer ring of neutron multiplying material 240. Axially disposed around the outer neutron multiplying alloy ring is a cladding 250, and may optionally include reflective material to reflect second generation neutrons back to the target to drive fission reactions. The reflector is preferably fabricated from beryllium, beryllium deuteride, beryllium tritride, or heavy water producing thermal spectrum neutrons and reflecting them back for capture or to drive a sub-critical chain reaction.

[0036] Channels 215, 225, 235, 245, may be disposed around and between each of the material layers and provided with circulated air or selected liquid metal or heavy water to cool each layer of the target assembly. Alternatively, in embodiments where cooling is either not desired or not needed, the channels may be eliminated and each material layer (ring or tube) disposed immediately upon and approximated to the underlying material layer (ring, tube, or rod).

[0037] FIG. 5 is a cross sectional end view in elevation of the central neutron generator cavity where the capsule attaches to an electronics package 270, a gamma generating alloy forming the cup 280, and selected target material 290.

[0038] As will be readily appreciated from the foregoing description of the essential structural elements comprising the compound target assembly, the inventive apparatus includes several unique design characteristics. There are three common novel elements to each embodiment of this innovation. The first novel element is the use of neutron multiplication in two or more volumes within the compound target assembly. These volumes are depicted in the drawings

as having a rod-in-tube configuration (power rod, or inner neutron multiplying rod **200**, and outer ring **240**), but they take a plate-on-plate configuration or a disk-on-disk configuration. In the rod-in-tube configuration, the “power rod” is the central neutron multiplying alloy rod or tube **200**, and the “power tube” is the outer neutron multiplying tube **240**. In addition to receiving thermal spectrum neutrons from the general reactor volume, the target material “sees” an optimized spectrum with a higher flux produced “locally” from the fissioning of the fissile material in the alloy in at least two locations within the device, preferably on both the inner and outer sides of the target isotope material. The same effect is produced by stacked plates or an assembly of disks.

**[0039]** The fission spectrum neutrons produced in the neutron multiplier will have a mean travel range of more than a centimeter in the solid alloys used for neutron conduction to the shaping alloy. Runaway heat or overheating is avoided, firstly by the use of thorium, which absorbs excess neutrons if the population of neutrons becomes too extensive at any point in time, and secondly by conventional cooling with supercritical carbon dioxide gas or water or a liquid metal coolant such as gallium, sodium, lithium or lead bismuth eutectic. The fissile material is preferably uranium-233, uranium-235, plutonium-239, or any optimized combination thereof. Plutonium is the least favored fissile material because of proliferation concerns. Fissile uranium can be denatured with uranium-238 to allay proliferation concerns, but if concentrations of uranium-233 must exceed twelve percent (12%) or concentrations of uranium-235 must exceed twenty percent (20%) to obtain the requisite spectrum, then anti-proliferative denaturing can be accomplished by the addition of uranium-232 and protactinium-231 to the power or driver rod, tube, plates or disks. Thorium is used as fertile material to replace uranium-238 where possible.

**[0040]** The fissile material comprises dispersed metallic particles or as ceramic, meaning the dispersed particles are nitride, boride, carbide, silicide of the fissile material selected. The preferred fertile material is thorium-232 oxide at approximately 85% of maximum density, desired because of its chemical stability, high temperature tolerance, and plenum volumes for fission gasses and fission products. This fertile oxide is dispersed in the alloys comprising silumen, an alloy of aluminum and silicon, vanadium, aluminum, tungsten, tantalum, molybdenum, zirconium, chromium, or with scattering materials like iron and nickel. This first metal matrix functions as a neutron multiplier and a receptacle to contain and retain fission products produced within it to keep the target material free of fission product contamination. Thus, the first metal matrix generates fission spectrum neutrons and traps the fission products.

**[0041]** The second element common to all of the embodiments is the spectrum shaping alloy. This material is disposed in the target assembly between the target isotope and the two neutron multiplying layers disposed on each side of the target isotope. The spectrum shaping alloy contains materials to soften the fission spectrum to a fast neutron spectrum, or to an intermediate or an epithermal neutron spectrum, as needed to promote the most efficient capture in the selected target material. As a safety feature, the resonances in the neutron absorption cross-sections of thorium assist to absorb neutrons when the temperature of the assembly is too hot.

**[0042]** The metal matrix for each embodiment is composed of either a nickel aluminide, zirconium aluminide, nickel-chromium-vanadium, or nickel-aluminum-zirconium-vana-

dium alloy that has been optimized to shape the neutron spectrum. Aluminum has a low parasitic neutron absorption cross-section, and nickel has a high elastic scattering cross-section to trap neutrons in the core.

**[0043]** Hydrogen atoms in the hydrides in a fast or an epithermal compound target assembly function to soften neutron spectrum to its optimal shape for transmutation of the desired end product. Further, deuterium can be used in place of hydrogen for some applications. Metals preferred in forming the intermetallic hydride or deuteride compounds include erbium, ytterbium-176, zirconium, vanadium, titanium. Additionally, various oxides are known to shape or tailor neutron spectra. These include zirconia, alumina, and lithium-7. Finally carbide compounds and nitrides disposed as homogeneously dispersed fine particles can be used with the hydrides to assist in slowing neutrons in aluminum alloys. All of the additives are fine powders that are homogeneously dispersed in the aluminum alloy.

**[0044]** The fissile alloy for the neutron multiplier is preferably clad in a zirconium alloy used in light water reactors or HT-9 so that the exterior surface of the compound target assembly is identical to or very similar to the cladding of the reactor into which the compound target assembly is placed in service. A preferred structural alloy is nickel aluminide, desirable because of its high thermal conductivity and strength at high temperature, and also because it generates photons of high enough energy to displace neutrons from target materials. Other structural alloys include aluminum vanadium mixed with zirconium.

**[0045]** As will be readily appreciated, there are numerous possible variations on the preferred embodiments.

**[0046]** One use for the device is in the economic production of molybdenum-99 for the technetium-99m market. In such an application, the compound target assembly may have the following characteristics.

**[0047]** First, the target material is molybdenum-98, having a purity of at least 99.9%. As illustrated in the drawings, it is deployed in a tube **220** disposed between the two spectrum shaping tubes **210**, **230**, which are, in turn, disposed between the central neutron multiplying rod (i.e., the “power rod”) **200** and the outer neutron multiplying tube **240**. The major capture resonances of molybdenum-98 are found in an energy range between 5 keV and  $5 \times 10^5$  ( $5 \times 10^5$  sup.5) keV.

**[0048]** The preferred alloy for shaping the spectrum is composed of aluminum zirconium holding very fine and homogeneously dispersed scattering particles, preferably of boron-11 carbide. Other materials may also be employed, as noted.

**[0049]** The inner tube of spectrum shaping alloy **210** is preferably 0.5 cm in thickness and fits over the center neutron multiplying rod **200**, which has a smaller outer diameter than the interior diameter of the inner tube of spectrum shaping alloy.

**[0050]** Next, the target material tube **220** is made of molybdenum-98 and is disposed over the inner neutron multiplying tube and the inner spectrum shaping tube. Its inner diameter is preferably 1.5 cm, and its exterior diameter is 2 cm. The molybdenum target metal is finely powdered, sintered in vanadium to fit in the dimensions of the exterior production tube.

**[0051]** The next tube is an outer tube of spectrum shaping alloy **230** that fits over the target isotope tube. Its inner diameter is 2.2 cm and its exterior diameter is 2.7 cm.

**[0052]** Disposed around the outer tube of spectrum shaping material is an outer neutron multiplier alloy tube **240**, prefer-



ably including homogeneously distributed fissile material in the alloy. It is a tube of 2.8 cm inner diameter and 3.5 exterior diameter.

**[0053]** Lastly, a cladding **250** is disposed around the outer neutron multiplier tube. This will generally be fabricated from the same material as the cladding material used to clad the fuel of the reactor in which the compound target assembly is placed for irradiation.

**[0054]** In use, the compound assembly is placed in the “rabbit” of a rabbit reactor system, and the fissile material commences fissioning. Heat is extracted through the metal matrix. In some configurations, the central “rod” must be embodied in a tube form so that it, too, can be cooled by circulated liquid metal or a compressed gas such as helium or carbon dioxide. The fissile fertile isotope-containing alloy is designed to trap fission products and gasses in thorium oxide dispersed throughout the material. Energetic neutrons depart this material, fission gasses and fission products are trapped and remain behind, and the neutrons enter the spectrum shaping alloy. Here the spectrum is shaped or tailored so that thermal neutrons are trapped or reflected back to the neutron multipliers and the fission spectrum neutrons lose a calculated percentage of their energy largely due to the scattering effects of the “additives” such as boron-11 carbide in the aluminum zirconium vanadium alloy. Thus, the shaped spectrum of neutrons enters the molybdenum-98 target from two proximate sources at the energies where capture is greatest, namely, at approximately 5 keV. The spectrum of the neutrons leaving the shaping alloy is, on average, slightly above the major resonance region of molybdenum-98, not far above  $5 \times 10^5$  ( $5 \times 10^{\text{sup.}5}$ ) electron volts. This energy is just above the major resonance energy levels of the molybdenum-98 nuclei that the neutrons will encounter through inside the target.

**[0055]** The target is irradiated for a duration of three half lives of molybdenum-99, or roughly eight and one quarter days, as one half life is 65.94 hours. When the production cycle is complete, sufficient quantities of isotopes will be present in the target powder, which can be placed inside a lead cow for delivery to a hospital. The material can be returned in a week or ten days for more irradiation to complete the supply cycle.

**[0056]** In addition, the power or driver rods can be subject to chemical separation, and some molybdenum-99 will be produced from the fission products.

**[0057]** Doppler effects caused by the thorium 232- or uranium-238 reduce the rate of fission as temperature increases above normal operating temperature, and this characteristic is positively exploited for safety reasons.

**[0058]** The procedure for the production of holmium-166 is less complicated than that for molybdenum-99, as holmium is mono-isotopic and does not need an isotopically separated target (natural holmium is 100% holmium-165). The major resonances are in the epithermal range with two peaks, one at one hundred electron volts (100 eV) and the other at near five electron volts (5 eV). Holmium-166 has a half life of 26.83 hours. Thus, three half lives of holmium-166 is approximately three days and six hours. This is the most effective irradiation time. In the production cycle, holmium is finely divided, compacted, and placed in a vanadium “can” between the spectrum shaping tubes, plates or disks, and the neutron multiplying driver tubes, plates or disks.

**[0059]** The procedure for the production of copper-64 and copper-67 is simpler still. Here the object is to expose the

precursors, zinc-64 and zinc-67, to a high flux fission spectrum. For this case, there is no need for spectrum shaping. Rather, the method involves increasing the high energy flux level to effectively transmute the zinc targets to produce the desired copper isotopes using an n,p reaction. For this embodiment the spectrum shaping alloy is superfluous; however, the multiplier alloys are required to act as a trap for fission products and gasses as well as to produce the fission spectrum neutrons. The expected irradiation time is different for each, being about three half lives, 36 hours for copper-64, and 7.5 days for copper-67.

**[0060]** The next isotope suitable for production in the inventive compound target assembly is rhenium-186. For this, rhenium-185 is the target. The major resonances are in the range of five electron volts (5 eV). This alloy is aluminum with zirconium hydride designed to slow the neutron spectrum down to almost thermal ranges. The expected irradiation time is eleven and one quarter days.

**[0061]** Yet another suitable isotope is tungsten-188. It is produced by two successive neutron captures starting with tungsten-186. Tungsten-186 is exposed to the fission spectrum neutrons that are tailored by zirconium hydride in matrix alloy of aluminum, vanadium and zirconium, such that the epithermal neutrons encountering the tungsten-186 and tungsten-187 are more likely to be captured and rhenium-188 is produced as a decay product of tungsten-188. The major resonances of tungsten-186 peak at around ten electron volts (10 eV). The shaping alloy will reduce the energy of the fission spectrum to around on hundred electron volts (100 eV) to allow for optimal simultaneous transmutation.

**[0062]** Another isotope is radium-223. The precursor isotope material is radium-226, which is exposed to epithermal neutrons to make actinium-227. Through several decays the actinium-227 produces radium-223. The neutron spectrum most conducive for transmutation is between one tenth of an electron volt ( $1/10$  eV) and one electron volt (1 eV). Here the alloy will be hydrided with zirconium hydride to thermalise the enhanced neutron flux to the correct energy so as to optimize production.

**[0063]** Many other isotopes may be produced, include, among others, lutetium-177, yttrium-90 and rhenium-188, primarily by neutron capture at target resonance energies. Also the same alloys can be applied to the treatment of fission products so that the long lived fission products can be transmuted to shorter lived isotopes or stable isotopes. Technetium-99 and iodine-129 can be converted to ruthenium-100 or xenon-130, respectively, by exposure to epithermal neutrons.

**[0064]** While the preferred embodiment is described as either a rod-in-tube configuration, or a plate-on-plate or disk-on-disk configuration, an alternate embodiment may make use of thin metal foils rather than rods and tubes, plates, disks, or cans. Even so, there remain the fundamental structural and operational components of the inventive apparatus—namely, the multiplier, shaper, and target layers—but in an alternative embodiment they may be provided in foil form. In such an embodiment, the foils are rolled together and disposed inside a tube of zirconium alloy cladding and sealed. The target is irradiated by fission spectrum neutrons whose spectrum is shaped by the spectrum shaping foil. This embodiment is much more simple to fabricate and irradiate, but it increases the chance that fission products will contaminate the target. When this embodiment is used, chemical separation techniques should be employed to gather useful fission products as well as the capture material product. The foil approach may

be the best for the treatment of fission products to transmute them to shorter lived or stable isotopes.

**[0065]** Summarily stated in the most essential and general terms, and as should be clear from the foregoing description, a target isotope or parent material is placed into close proximity with a neutron multiplying alloy containing fissioning material. The fissioning material may be uranium-233, uranium-235 or plutonium-239, americium-243, or curium-245, or fissioning material, either alone or in any combination thereof, and provided in oxide, nitride, boride or carbide form (i.e. ceramic form as homogeneously dispersed particles) or in metallic form as homogeneously dispersed particles. These are placed in a matrix metal of neutron “conducting” alloy composed of aluminum or silumen (a compound of silicon and aluminum) with engineered amounts of zirconium, vanadium, chromium, nickel, molybdenum or tungsten as an alloy composed of them designed for a particular composite target module, taking into account the properties of the fissile material, the properties of the fertile fissile material (such as thorium), and the properties of the target so that the neutrons escaping from the alloy will have the energy of the resonance integral of the target material. This vastly increases the probability of neutron capture by the target isotope and provides an efficient and economically viable transmutation of the target isotopes into useful medical or commercial isotopes. This also provides for the economic transmutation of fission products to shorter lived isotopes or stable isotopes.

**[0066]** The neutron multiplying alloys function as a filter, keeping the fission products, including fission gasses, in the inert metal matrix, letting the fission spectrum neutrons out, for passage through a second material engineered to function to shape or moderate the fission spectrum to promote the spectrum shaping or tailoring effects so that the neutrons entering the target will have an energy above the target isotopes resonance integral.

**[0067]** Depending on the isotope to be produced, a different alloy is used to shape the output from the neutron multiplier so that each target isotope is irradiated by neutrons having a tailored neutron spectrum for neutron capture at target resonance energies. The geometry and the composition of each shaping or tailoring alloy permits neutrons from the multiplier to develop a useful energy spectrum upon departure from the shaping alloy for entry into the targets. The synthesized neutron energy spectra are designed to improve probabilities of neutron capture in the selected target material either for isotope production or fission product disposition.

**[0068]** Spectrum tailoring or shaping provides a very efficient neutron capture method of isotope manufacture, wherein neutrons are added to the target, and for the gamma neutron reactions where neutrons are ejected from the target. This shaping is done with the novel alloys disclosed herein. The precursor or target material is generally an elemental isotope or an enriched stable isotope. The target isotope captures neutrons generally most efficiently at resonance energies of the target isotopes. For each of the target materials selected, the capture energies or resonance energies are a unique property of the isotope of the target.

**[0069]** The product isotopes discussed above are examples of medical isotopes that have high value for the treatment and diagnosis of human diseases and are in short supply due to inefficient or unavailable means and methods of production. The approach revealed in this application supplements existing means to fabricate these beneficial materials.

**[0070]** Having fully described several embodiments of the present invention, many other equivalents and alternative embodiments will be apparent to those skilled in the art. These and other equivalents and alternatives are intended to be included within the scope of the present invention.

What is claimed as invention is:

1. A compound target assembly for producing medical and commercial isotopes, comprising:

a charged particle source for providing a beam of accelerated charged particles;

a neutron multiplier alloy spaced apart from said charged particle source and at which a beam of charged particles is directed when in use;

a target precursor isotope; and

a spectrum shaping alloy disposed between said neutron multiplier alloy and said target isotope.

2. The apparatus of claim 1, wherein said charged particle source is a 10 milliamp electron beam which directs electrons accelerated to 16 to 28 MeV at said neutron multiplier alloy to produce a rich flux of fission spectrum neutrons, and wherein said spectrum shaping alloy shape the energy spectrum of the fission spectrum neutrons to increase the probability of neutron capture by said target precursor isotope.

3. The apparatus of claim 1, including first and second volumes of neutron multiplier alloy, said first neutron multiplier alloy disposed in a centrally disposed power rod, and said second neutron multiplier alloy disposed in an outer tube.

4. The apparatus of claim 1, including first and second volumes of neutron multiplier alloy and first and second volumes of neutron spectrum shaping alloy.

5. The apparatus of claim 4, wherein said apparatus is a substantially cylindrical capsule having a rod-in-tube configuration.

6. The apparatus of claim 5, wherein said first neutron multiplier volume is substantially a rod disposed in the general center of the cylinder, said first neutron spectrum shaping alloy is disposed in a tube axially surrounding said first neutron multiplier volume, said target isotope is disposed in a tube axially surrounding said first neutron spectrum shaping volume, said second neutron spectrum shaping volume is a tube axially surrounding said target isotope, and said second neutron multiplier volume is disposed a tube axially surrounding said second neutron spectrum shaper.

7. The apparatus of claim 6, further including one or more cooling channels disposed between one or more of said first and second neutron multiplier alloy volumes, said first and second neutron spectrum shaping alloys, and said target isotope.

8. The apparatus of claim 7, further including a reflector axially disposed around said second neutron multiplier volume.

9. The apparatus of claim 1, wherein said apparatus has a plate-on-plate configuration.

10. The apparatus of claim 1, further including one or more channels for circulating a coolant through said apparatus.

11. The apparatus of claim 10, wherein said coolant is selected from the group consisting of supercritical carbon dioxide gas, water, and liquid metal.

12. The apparatus of claim 11, wherein said liquid metal is selected from the group consisting of gallium, sodium, lithium and lead bismuth eutectic.

13. The apparatus of claim 1, wherein said neutron multiplier alloy includes thorium to absorb excess neutrons and to prevent overheating.

**14.** The apparatus of claim **1**, wherein neutron multiplier alloy is a metal matrix that includes fissile material.

**15.** The apparatus of claim **14**, wherein said fissile material is uranium-233, uranium-235, plutonium-239, and any combination thereof.

**16.** The apparatus of claim **14**, wherein said fissile material comprises dispersed metallic particles in the form of nitrides, borides, carbides, silicides, and any combination thereof.

**17.** The apparatus of claim **1**, wherein said neutron multiplier alloy includes a fertile material.

**18.** The apparatus of claim **16**, wherein said fertile material is a fertile oxide.

**19.** The apparatus of claim **18**, wherein said fertile oxide is thorium-232 oxide.

**20.** The apparatus of claim **18**, wherein said fertile oxide is dispersed in an alloy selected from the group consisting of silumen, vanadium, aluminum, tungsten, tantalum, molybdenum, zirconium, chromium, and may further including scattering materials.

**21.** The apparatus of claim **20**, wherein said scattering materials comprise iron and nickel.

**22.** The apparatus of claim **1**, wherein said spectrum shaping alloy contains materials for softening fission spectrum neutrons to a fast, intermediate, or epithermal neutron spectrum, so as to increase the probability of neutron capture in said target isotope material.

**23.** The apparatus of claim **22**, wherein said spectrum shaping alloy is a metal matrix fabricated from material selected from the group consisting of nickel aluminide, zirconium aluminide, nickel-chromium-vanadium, and nickel-aluminum-zirconium-vanadium.

**24.** The apparatus of claim **23**, wherein said spectrum shaping alloy includes neutron slowing material selected from the group consisting of oxides, hydrides, deuterides, and carbides.

**25.** The apparatus of claim **24**, wherein said neutron slowing materials are hydride and deuteride compounds formed from elements selected from the group consisting of erbium, ytterbium-176, zirconium, vanadium, and titanium, and said oxides are compounds formed from the group consisting of zirconia, alumina, and lithia-7.

**26.** The apparatus of claim **24**, wherein said carbides is boron-11 carbide dispersed in an aluminum zirconium vanadium alloy.

**27.** The apparatus of claim **1**, wherein said neutron spectrum shaping alloy is an aluminium alloy including hydrides and homogeneously dispersed particles of either carbide compounds or nitrides to assist neutron slowing.

**28.** The apparatus of claim **1**, wherein said compound target assembly is clad in a zirconium alloy.

**29.** A compact compound target assembly, comprising:

a target isotope disposed in close proximity with a neutron multiplying alloy, said neutron multiplying material containing fissioning material.

**30.** The apparatus of claim **29**, wherein said fissioning material is an oxide, nitride, boride, or carbide compound formed with an isotope selected from the group consisting of uranium-233, uranium-235, plutonium-239, americium-243, curium-245, and any combination thereof,

**31.** The apparatus of claim **30**, wherein said fissioning material is in ceramic form as homogeneously dispersed particles.

**32.** The apparatus of claim **31**, wherein said fissioning material is in metallic form as homogeneously dispersed particles placed in a matrix metal of neutron conducting alloy.

**33.** The apparatus of claim **32**, wherein said neutron conducting alloy is fabricated from aluminum or silumen and includes material selected from the group consisting of zirconium, vanadium, chromium, nickel, molybdenum, tungsten, and any combination thereof.

\* \* \* \* \*