Sept. 26, 1972 J. W. BLUE ET AL 3,694,313

PRODUCTION OF HIGH PURITY ^{123}I

Filed Oct. 2, 1969 2 Sheets-Sheet 1

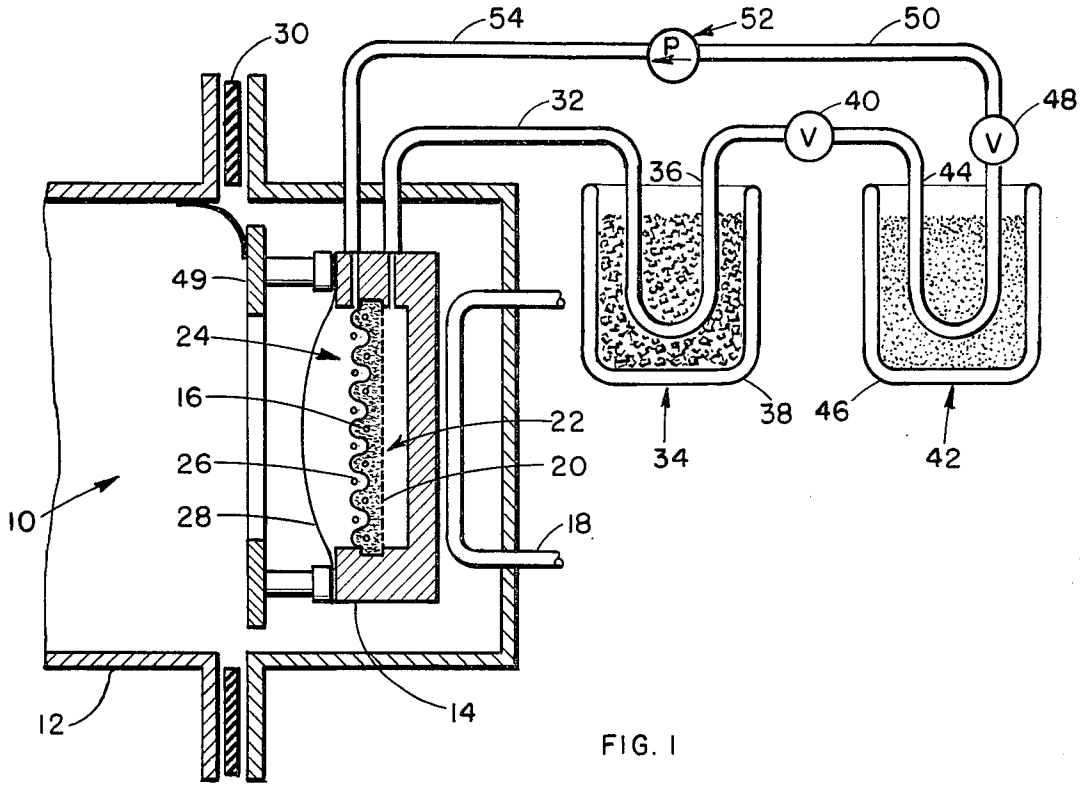


FIG. 1

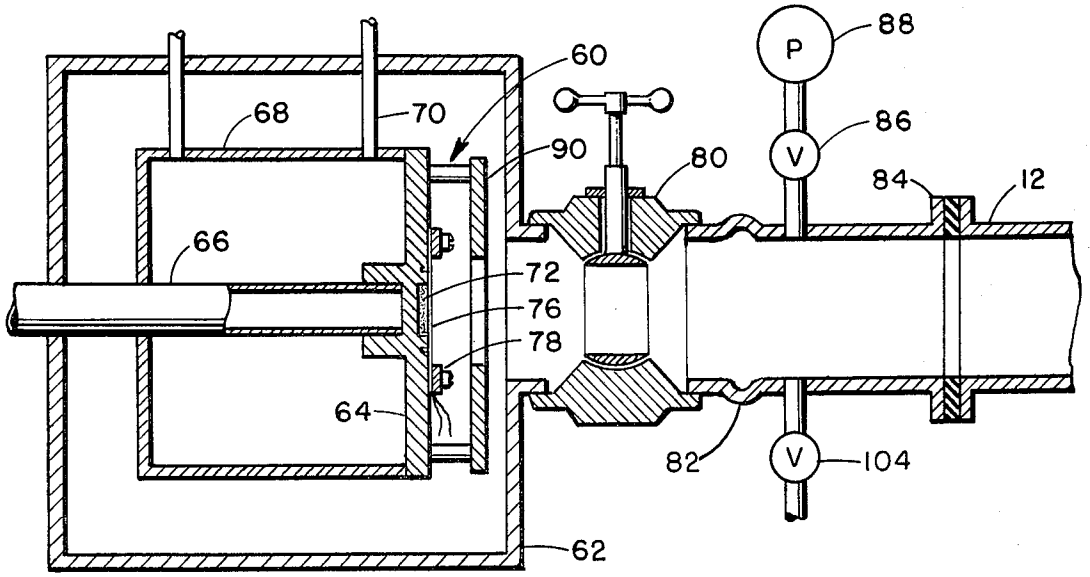


FIG. 2

INVENTORS

JAMES W. BLUE
WAYNE R. SMITH
VINCENT J. SODD

BY

Glenn & Coon
Irene E. Shook ATTORNEYS

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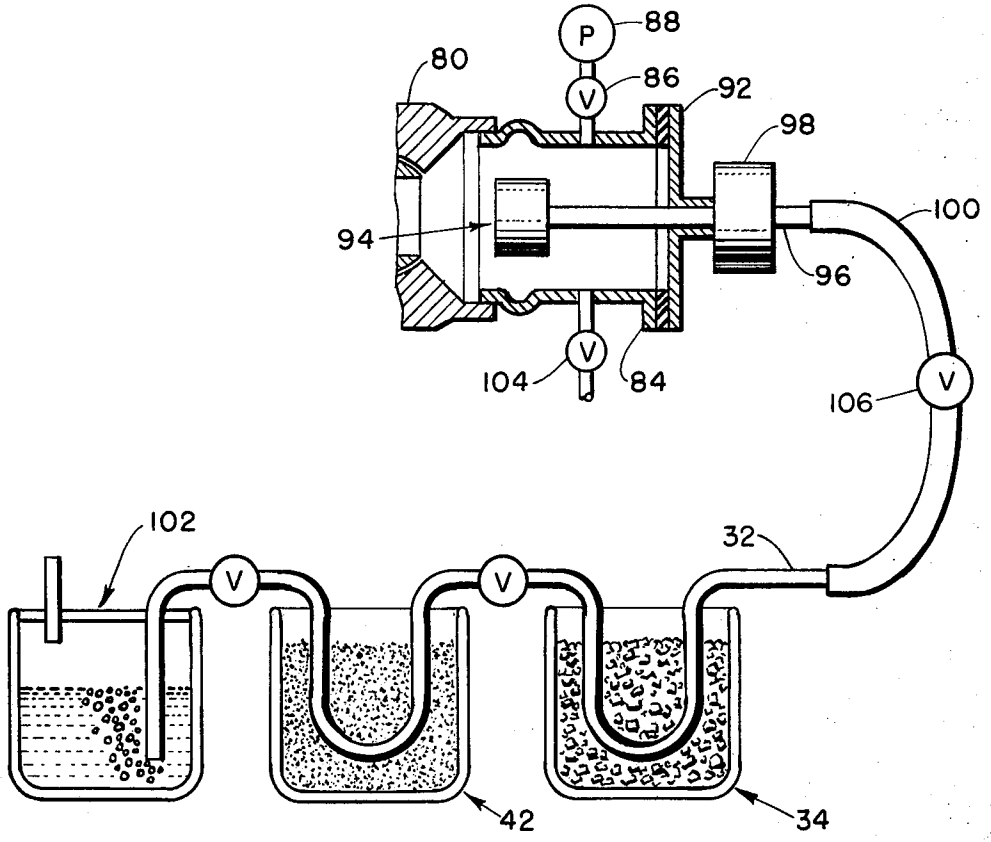


FIG. 3

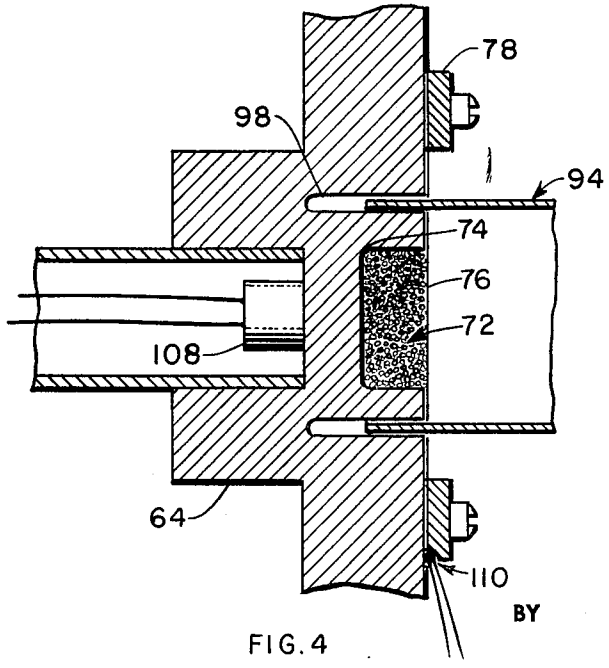


FIG. 4

INVENTORS
JAMES W. BLUE
WAYNE R. SMITH
VINCENT J. SODD
BY
Gene E. Shook ATTORNEYS

United States Patent Office 3,694,313 Patented Sept. 26, 1972

3,694,313

PRODUCTION OF HIGH PURITY ^{123}I

James W. Blue, Bay Village, Wayne R. Smith, Columbia Station, and Vincent J. Sodd, Cincinnati, Ohio, assignors to the United States of America as represented by the Administrator of the National Aeronautics and Space Administration

Filed Oct. 2, 1969, Ser. No. 863,280
Int. Cl. G21g 1/00
U.S. Cl. 176—11 1 Claim

ABSTRACT OF THE DISCLOSURE

Bombarding a tellurium target with a beam from an accelerator produces ^{123}Xe which is carried away by a flowing gas stream. Contaminants are removed from the gas, and the remaining xenon decays to ^{123}I which is ready for use as a radiopharmaceutical in which low radiation exposure is desired as in diagnostic studies.

ORIGIN OF THE INVENTION

The invention described herein was made by employees of the United States Government and may be manufactured and used by or for the Government for governmental purposes without the payment of any royalties thereon or therefor.

BACKGROUND OF THE INVENTION

This invention is concerned with the production of high purity radioiodine for thyroid measurements and as a general radionuclide. The invention is particularly directed to a method of producing ^{123}I by bombarding ^{122}Te or ^{123}Te with a beam that is within the energy limitations of a compact cyclotron.

Radioactive iodine is used for medical diagnostic studies. The isotope ^{131}I has been used for this purpose because of its availability. The substitution of ^{123}I for ^{131}I has been proposed in studies where the amount of radiation exposure to a patient is of prime concern. Because of the shorter half-life and the decay by electron capture, the radiation exposure received by the patient from ^{123}I is about $\frac{1}{40}$ that of an equal amount of ^{131}I. Another factor is that the gamma ray energy of ^{123}I is 159 kev. compared to 364 kev. of ^{131}I. Collimators operate more effectively with this lower energy, and they are less bulky.

It has been proposed that ^{123}I be produced by several methods utilizing ^{122}Te (α, 3n); ^{121}Sb (α, 2n); ^{122}Te (p, n); and ^{122}Te (He^3, 2n) reactions. However, the contaminants produced from these reactions prevent the full benefit of the ^{123}I from being achieved. The advantage of lower radiation exposure with the use of ^{123}I is only fully realized if the contamination by other radioiodines is small. By way of example, a 1% contamination of ^{123}I with ^{124}I doubles the patient radiation exposure.

Most of the ^{123}I used for nuclear applications has been made by two methods using large cyclotrons to produce ^{123}Te (p, n) and ^{121}Sb (α, 2n) reactions. While these two methods give yields of ^{123}I which are adequate for the diagnostic studies, the processes are expensive because of the cost of the large cyclotrons. Also, contamination in the amount of 0.5 to 0.9% of ^{124}I is encountered. Another problem is that both methods require dissolution of the target material in sulphuric acid and subsequent distillation of ^{123}I into NaOH. Further, the target material must be chemically recovered, and this increases the production cost.

SUMMARY OF THE INVENTION

These problems have been solved by the present invention which utilizes a compact cyclotron to bombard

an enriched ^{122}Te or ^{123}Te target with He^3 of energy equal to 30 mev. or less to produce ^{123}Xe according to a ^{122}Te (He^3, 2n), ^{123}Xe or ^{123}Te (He^3, 3n) ^{123}Xe reaction. The ^{123}Xe is transported from the target in a gas stream to low temperature traps where the undesirable iodine contaminants are removed. The xenon is held for a period of time sufficient for it to decay to ^{123}I.

OBJECTS OF THE INVENTION

It is, therefore, an object of the invention to produce ^{123}I having sufficient purity for use as a radiopharmaceutical.

Another object of the invention is to provide a method of making high purity ^{123}I using a compact cyclotron without the dissolution of the target together with the chemical separation of ^{123}I and the subsequent chemical recovery of the target.

A further object of the invention is to provide a reusable generator for producing large amount of ^{123}I wherein ^{123}Xe is physically separated from a tellurium target.

These and other objects of the invention will be apparent from the specification which follows and from the drawings wherein like numerals are used throughout to identify like parts.

DESCRIPTION OF THE DRAWINGS

FIG. 1 is a schematic view of a generator constructed in accordance with the invention for producing radioactive iodine;

FIG. 2 is a schematic view of apparatus for producing radioactive iodine by a cyclic method in the part of the cycle in which the target material is being bombarded;

FIG. 3 is a schematic view of a portion of the apparatus shown in FIG. 2 connected to cold traps; and

FIG. 4 is an enlarged partial view of a section of the apparatus shown in FIG. 2.

DESCRIPTION OF THE PREFERRED EMBODIMENT

Referring now to FIG. 1 a target assembly **10** constructed in accordance with the present invention is mounted in the beam duct **12** of an accelerator such as a 60-inch cyclotron. The target assembly **10** utilizes a cylindrical housing **14** for containing a target material **16**. The housing **14** is preferably of aluminum, and its temperature is controlled by circulating water in a cooling system **18**.

A porous plate **20** supports the target material **16** and separates the housing **14** into a pair of chambers **22** and **24**. The target material **16** is in the form of a powder which is preferably held against the porous plate **20** by a screen **26**.

A thin metal foil window **28** separates the cyclotron vacuum system from the front target chamber **24**. A window **28** of 7 mg./cm.^{2} aluminum has been satisfactory. An insulator **30** extending about the periphery of the cyclotron beam duct **12** electrically isolates the target assembly **10** from the cyclotron.

A tube **32** connects the chamber **22** to a cold trap **34**. Tygon tubing has been satisfactory. The trap **34** comprises a U-tube **36** immersed in a coolant in an insulated container **38**. A ¼ inch copper U-tube surrounded by solid CO_2 in a Dewar has been satisfactory. The Dry Ice maintains the trap **34** at a temperature of —79° C.

A valve **40** connects the Dry Ice trap **34** to a second cold trap **42**. A ¼ inch copper U-tube **44** immersed in liquid nitrogen in a Dewar **46** has been satisfactory. The liquid nitrogen maintains the trap **42** at a temperature of —196° C.

A valve **48** connects the liquid nitrogen trap **42** to a tube **50** that enters a self-contained gas pump **52**. The chamber **24** is placed in communication with the pump **52** through a tube **54**.

In operation, target material **16** of enriched tellurium powder is spread uniformly on the porous plate **20** to form a 20 to 90 mg./cm.^{2} layer. The retaining screen **26** is placed in contact with the tellurium powder, and the metal foil window **28** is mounted on the housing **14** to close the chamber **24**.

A beam of helium particles of energy equal to about 40 mev. or less from the cyclotron beam duct **12** passes through a carbon collimator **49**. The beam penetrates the foil window **28** and bombards the tellurium target. This bombardment of the tellurium produces xenon in accordance with the reactions ^{122}Te (α, 3n); ^{122}Te (He^3, 2n); or ^{123}Te (He^3, 3n).

A flow of helium gas is passed through the tellurium powder and porous plate **20** to carry away the ^{123}Xe into the Dry Ice trap **34** and then into the liquid nitrogen trap **42**. By utilizing a closed loop system as shown in FIG. 1 a gas flow rate may be used that both transports the xenon and cools the powder target material while conserving the helium gas.

The conservation of the helium gas is not an important consideration when very low flow rates are used. In such cases a single pass system of the type shown in FIG. 3 is used. This arrangement eliminates the pump, and no evacuation of the system is required because the single pass system can be flushed. However, with such low flow rates no cooling of the target is realized, and the gas is only utilized to carry the xenon to the cold traps.

It will also be appreciated that a high flow rate in the closed system may be sufficient to maintain the tellurium in contact with the porous plate **20** without using the screen **26**. However, such high flow rates may carry impurities through the trap **34** into the trap **42**.

The helium carries the xenon from the target assembly **10** through the tube **32** into the Dry Ice trap **34**. Contaminants with freezing points higher than —78° C. are frozen out of the gas stream. These contaminants cling to the walls of the cold trap **34**. Among the contaminants removed in the trap **34** are the undesirable ^{124}I, ^{125}I, and ^{126}I.

The helium gas flow then carries the xenon into the liquid nitrogen trap **42** where the xenon and other products with freezing points between —79° C. and —196° C. are frozen out. The helium gas is then recirculated through the pump **52** and lines **50** and **54** back to the target assembly **10**.

After bombardment the liquid nitrogen trap is sealed by closing the valves **48** and **40**. The U-tube **44** is removed from the liquid nitrogen in the container **46** and set aside until the ^{123}I ingrowth is optimized. The 2.1-hour ^{123}Xe decays to ^{123}I in about four to eight hours. This time may be used to transport the ^{123}I from the cyclotron to a laboratory.

After waiting for the ^{123}Xe to decay, the U-tube **44** is flushed with helium gas to remove remaining ^{123}Xe and ^{125}Xe. The trap is rinsed with dilute NaOH and the solution is counted to ascertain the presence of ^{124}I. The ^{123}I is placed in a suitable chemical form for its ultimate medical use.

Direct tagging of organic molecules is possible in the trap **42**. By way of example, macroaggregated serum albumen has been tagged with ^{123}I by merely contacting a solution with the trap **42**. The bulk gas temperature at a helium flow rate of five standard liters per minute exiting a trap 50 centimeters long and 3 millimeters in diameter is approximately the temperature of the wall of the trap. With such a flow rate the helium would cool the target material **16** and the gas temperature would be lowered enough to freeze the radio xenon. However, at such flow rates the radioiodines do not stick to the wall of the U-tube **36**, and the amount of ^{124}I in the liquid nitrogen trap **42** is excessive. At the five liter per minute flow rate the gas is not in the Dry-Ice trap **34** long enough to insure complete radioiodine diffusion to the wall of the U-tube **38**. This results in low trap efficiency.

Reducing the flow rate to 0.02 liter per minute increases the efficiency of the Dry Ice trap **34** to about 100%. However, no cooling of the target material **16** is realized.

A flow rate of 10 liters per minute is found to be adequate to maintain the tellurium powder target material **16** in contact with the porous plate **20** without using a retaining screen **26**. This flow rate is also adequate to cool the target material **16** sufficiently to avoid deterioration in the beam. However, this flow rate forces many contaminants from the Dry Ice trap **34** into the liquid nitrogen trap **42** and baffling is required in the trap **34**.

The generator shown in FIG. 1 has an advantage in that the yield per hour of bombardment is nearly independent of bombardment time. Thus, in utilizing a small cyclotron for the production of large amounts of ^{123}I, the length of bombardment time would be determined from schedule consideration or a minimization of trap changes rather than from optimizing the yield.

DESCRIPTION OF AN ALTERNATE EMBODIMENT

A cyclic system for producing ^{123}I is shown in FIG. 2. In this apparatus the tellurium is bombarded for a definite period of time and kept cold during this period so that no ^{123}Xe escapes. The bombardment is terminated, and the tellurium target is heated to a temperature sufficient to remove all the ^{123}Xe. Provision is made for a controlled heating cycle so that all of the xenon is diffused from the tellurium at a temperature low enough to insure that none of this tellurium is vaporized and lost from the system. The same target can be used for successive bombardments by repeating the cycle.

The cyclic method utilizes a target assembly **60** mounted in a housing **62**. The target assembly **60** includes a plate **64** on the end of a tubular member **66**. The plate **64** is preferably copper and forms the end of a chamber **68** which is kept filled with liquid nitrogen by an automatic filling system **70**.

Target material **72** in the form of tellurium powder is contained in a small depression **74** in the plate **64**, and aluminum foil cover **76** is mounted over the depression **74** by a ring **78**. A ball valve **80** is mounted on the housing **62** adjacent the target assembly **60**. A tubular passage **82** extends from the opposite side of the valve **80**. This passage is mounted on the cyclotron beam duct **12** by a flange **84**.

In operation, the system is evacuated by opening the valve **86** connected to a vacuum pump **88**. A beam from the duct **12** passes through the passage **82** and valve **80** toward the target assembly **60**. After passing through a carbon collimator **90** the beam strikes the target material **72**.

At the end of the bombardment period the ball valve **80** is closed. The target assembly **60** is kept at high vacuum conditions within the housing **62** while the assembly is removed from the cyclotron beam duct **12**.

A plate **92** is mounted on the flange **84** as shown in FIG. 3. A foil cutter **94** is mounted on a tubular handle **96** that extends through the plate **92**. The space in the passage **82** between the valve **80** and the plate **92** is evacuated by opening the valve **86** to the vacuum pump **88**.

The ball valve **80** is then opened, and the tubular handle **96** is pushed through a sliding seal **98** until the aluminum foil cover **76** is cut in the manner shown in FIG. 4. When the foil cutter **94** is fully inserted its end is received in a circular groove **98** at the bottom of the depression **74** in the plate **64** as shown in FIG. 4.

The opposite end of the tubular handle **96** is connected to a tube **100** for placing the target assembly in communication with cold traps **34** and **42** through a tube **32** in a manner previously described in connection with

FIG. 1. This embodiment does not use a recirculating system. The system is vented through an oil bubbler **102**.

With the cutter **94** fully inserted as shown in FIG. 4 helium gas is slowly admitted to the system by opening a valve **104** that is connected to a suitable gas supply. When the pressure reaches the desired level, preferably one atmosphere, valve means **106**, such as a clamp, on the flexible tube **100** is opened. The helium flows through the valve **80** and around the end of the cutter **94** in the groove **98** to the tubular handle **96**. This gas carries the xenon to the cold traps **34** and **42** in the manner previously described.

The helium flow between the cutting edge of the cutter **94** and the groove **98** is then adjusted to a low rate, such as about 20 milliliters per minute. Power is supplied to a heater **108** that is inserted in the tubular member **66** in the liquid nitrogen container **68**. The heater **108** is in contact with the back of the plate **64** to heat the tellurium powder **72**. The temperature of the plate **64** is monitored by a thermocouple **110**.

It will be appreciated that the ^{123}Xe produced during the bombardment in the cyclic system shown in FIGS. 2 and 3 is not effective in producing ^{123}I if it decays in the target material before it is removed in the heating cycle. The length of the radiation should not exceed two hours followed by heating.

We claim:

1. A method of producing high purity radioactive ^{123}I with a compact cyclotron as a source of a He^3 beam at an energy up to 30 mev. comprising the steps of

mounting a target in powder form of an isotope from the group consisting of ^{122}Te and ^{123}Te in a chamber, mounting said chamber on said cyclotron, evacuating said chamber, bombarding said target with said He^3 beam to produce ^{123}Xe for a period of up to two hours, cooling said target with liquid nitrogen during bombardment for preventing escape of ^{123}Xe, sealing said target after bombardment, removing said sealed target from said cyclotron, connecting a source of helium gas to said target, heating said target to evaporate said ^{123}Xe, injecting helium into said chamber to raise the pressure of same to one atmosphere,

carrying the ^{123}Xe from the target in said helium flowing at a low velocity at a rate of about 0.02 liter per minute,

passing said helium gas through a Dry Ice cold trap to remove iodine contaminants therefrom, subsequently passing said helium gas through a liquid nitrogen cold trap to remove ^{123}Xe therefrom, subsequently passing said helium gas through an oil bubbler and then venting said helium gas to atmosphere, holding the ^{123}Xe in said liquid nitrogen trap for a period of time sufficient for it to decay to ^{123}I.

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BENJAMIN R. PADGETT, Primary Examiner

H. E. BEHREND, Assistant Examiner

U.S. Cl. X.R.

176—16; 23—312