March 3, 1953 E. B. MILLER 2,630,375

METHOD OF CATALYTIC DESULFURIZATION OF GASES

Filed March 12, 1951 10 Sheets-Sheet 1

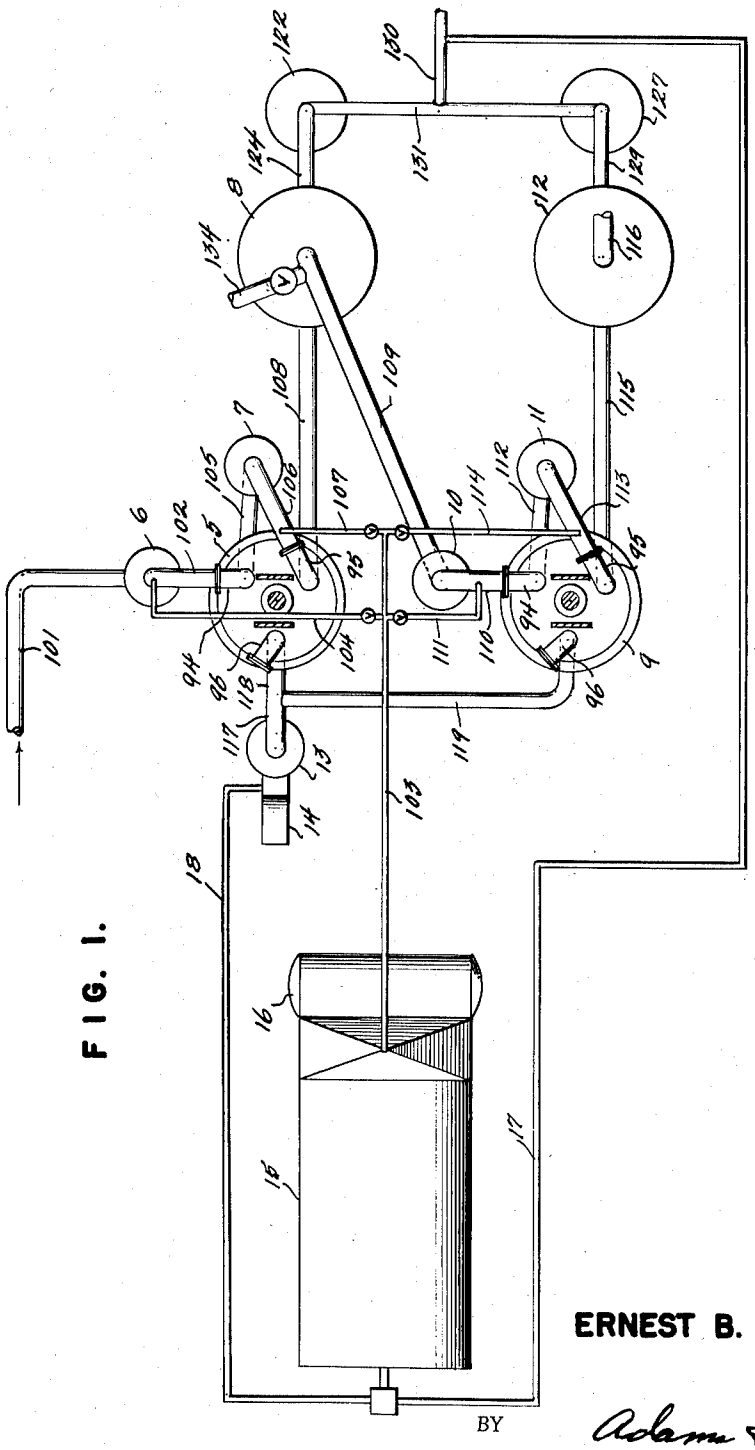


FIG. 1.

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METHOD OF CATALYTIC DESULFURIZATION OF GASES

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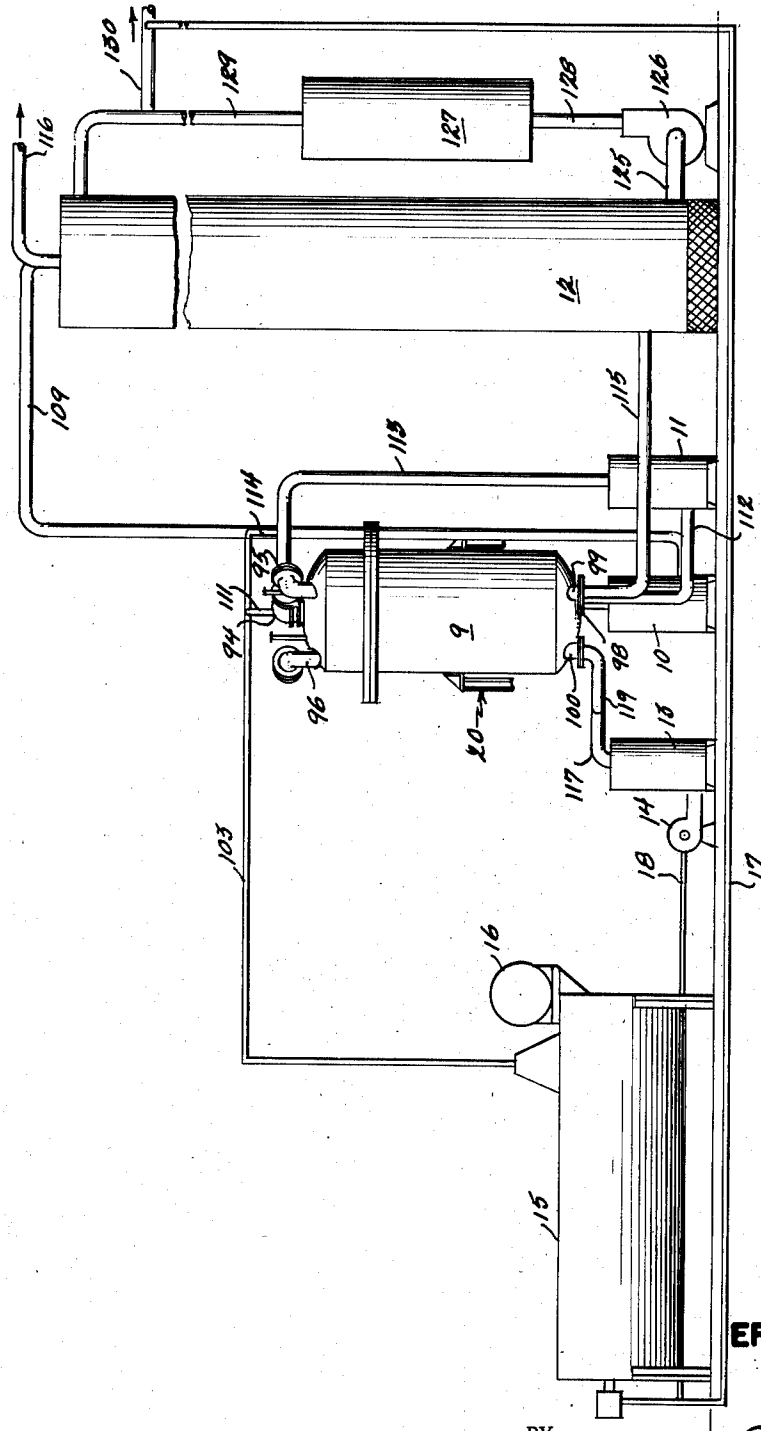


FIG. 2.

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METHOD OF CATALYTIC DESULFURIZATION OF GASES

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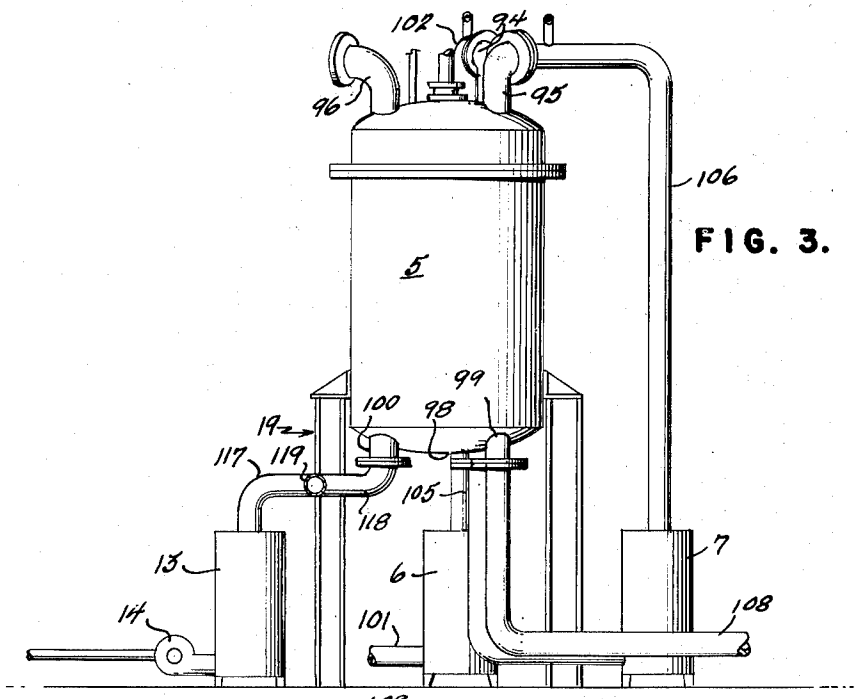


FIG. 3.

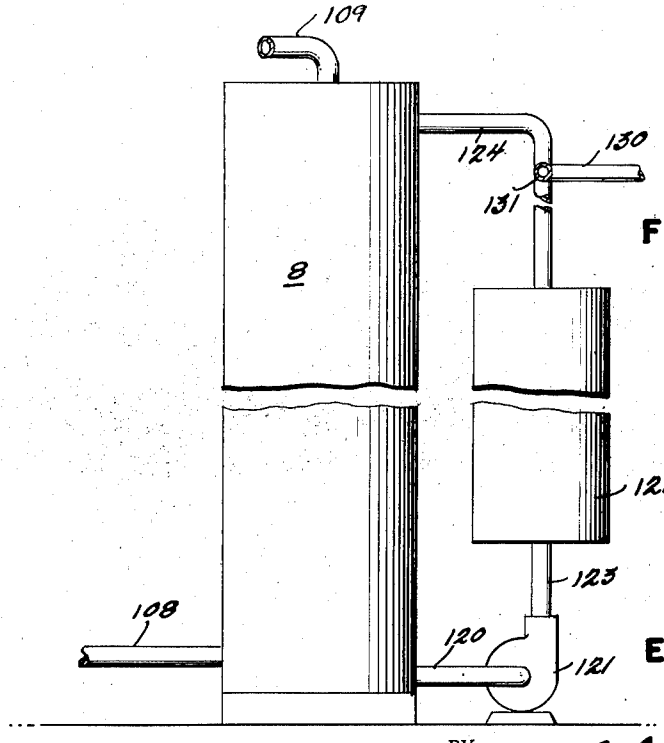


FIG. 4.

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March 3, 1953 E. B. MILLER 2,630,375

METHOD OF CATALYTIC DESULFURIZATION OF GASES

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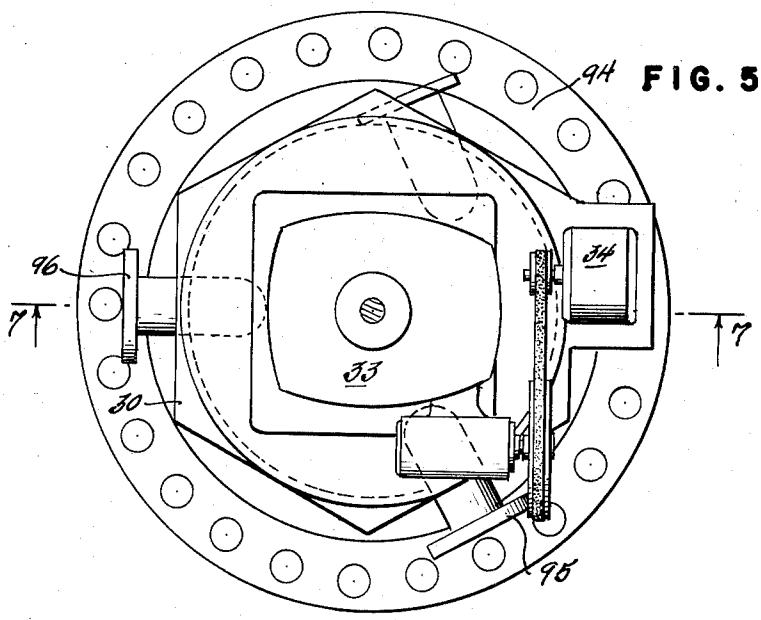


FIG. 5.

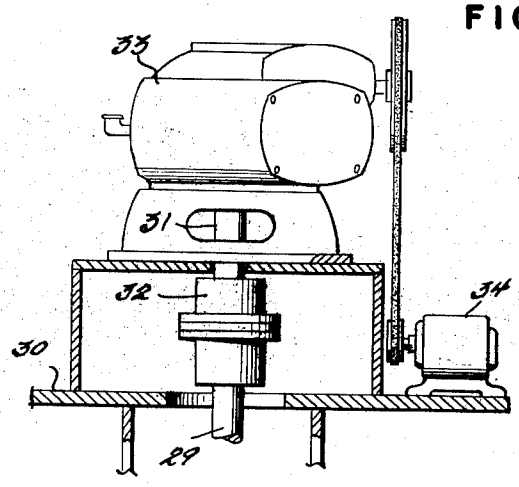


FIG. 6.

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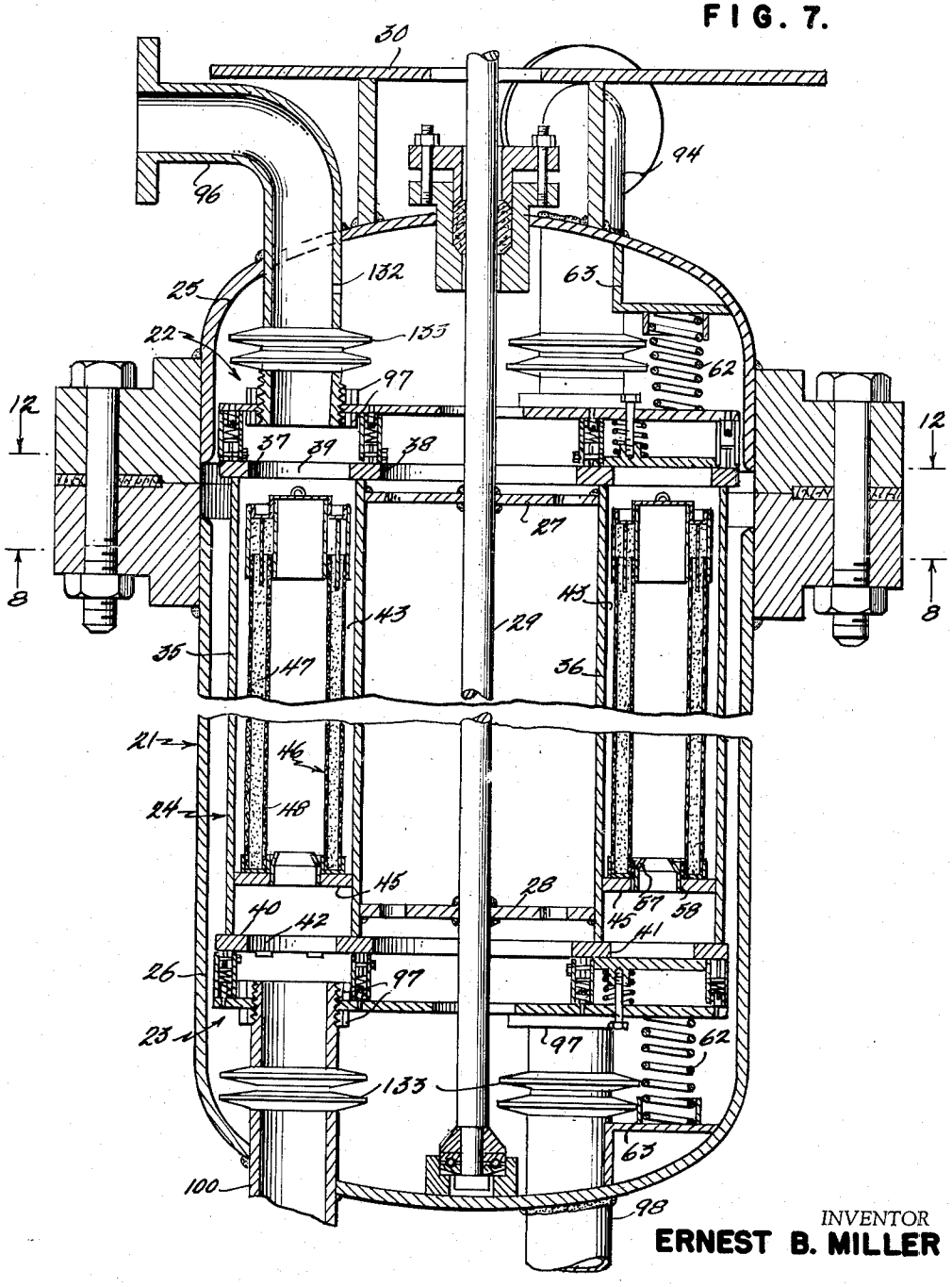
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METHOD OF CATALYTIC DESULFURIZATION OF GASES

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FIG. 7.



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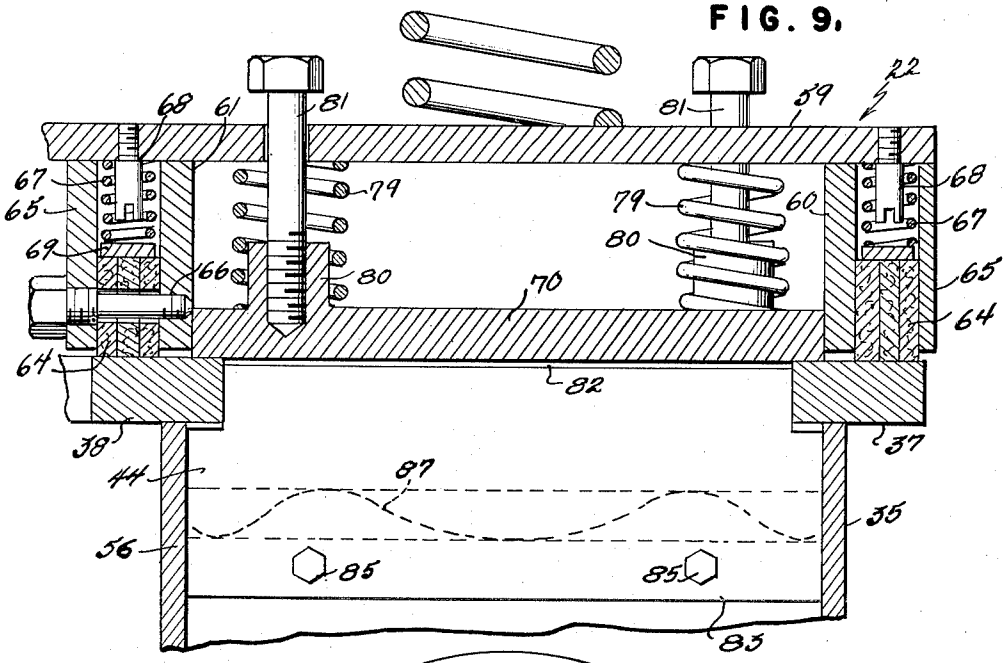
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METHOD OF CATALYTIC DESULFURIZATION OF GASES

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FIG. 9.



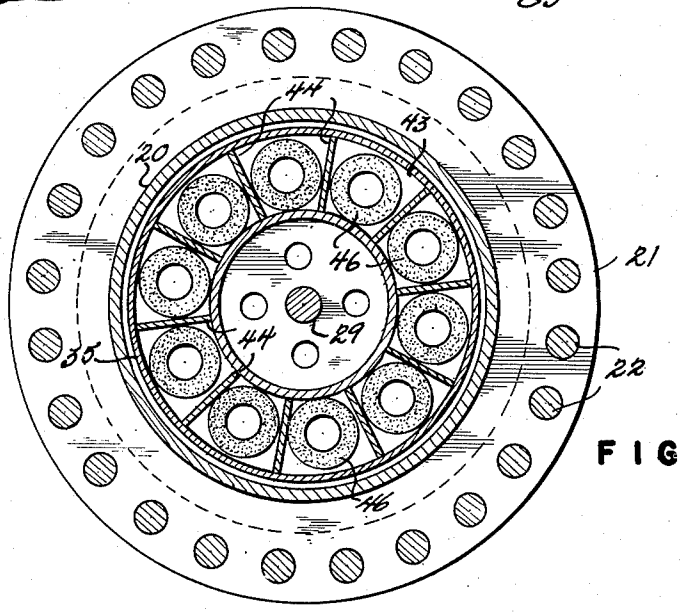


FIG. 8.

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METHOD OF CATALYTIC DESULFURIZATION OF GASES

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FIG. 10.

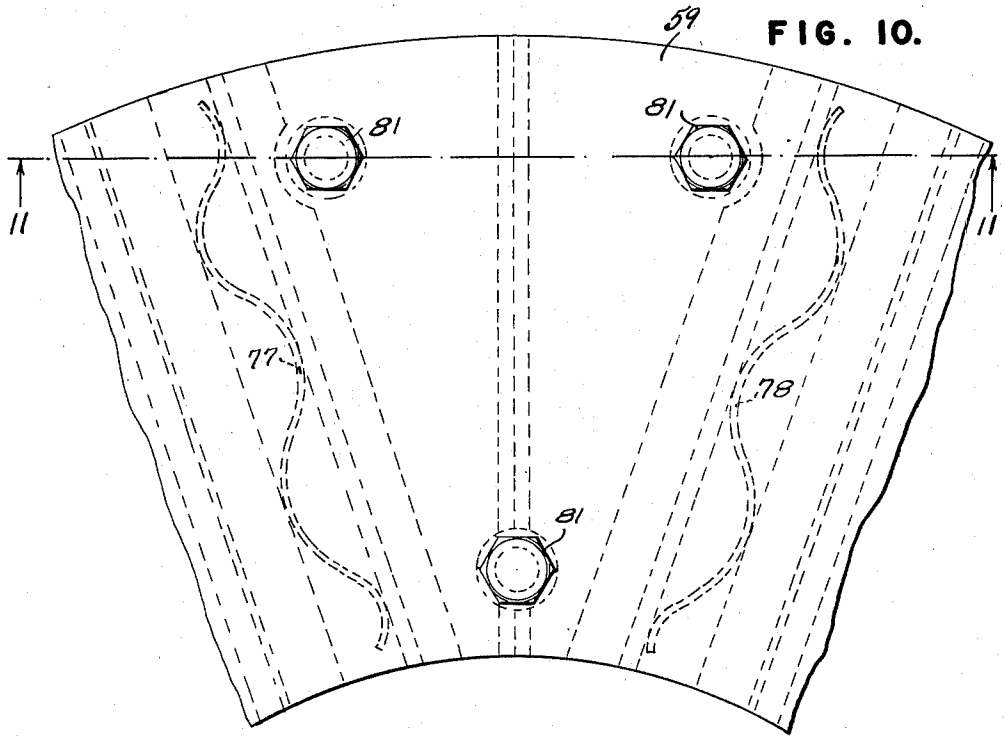
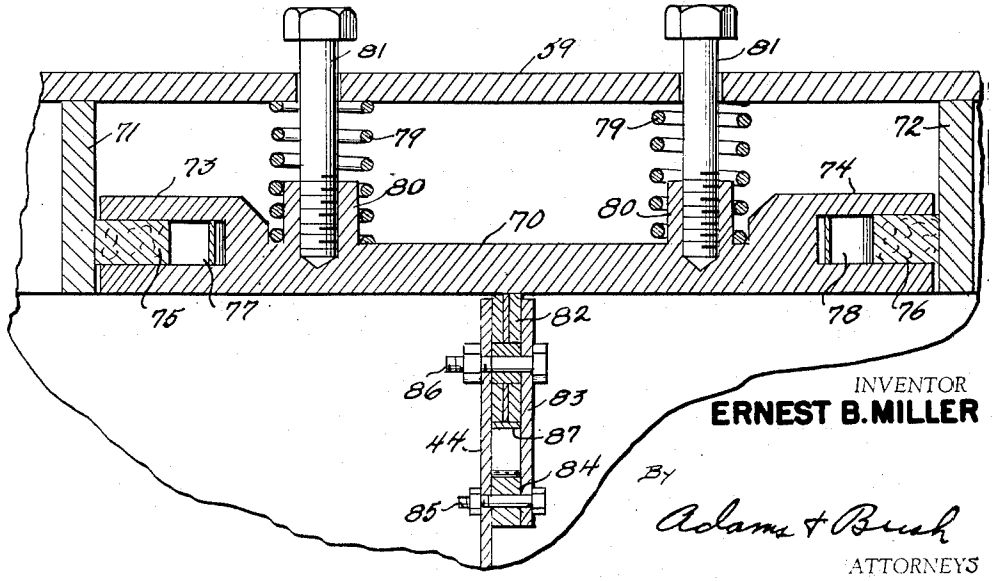


FIG. 11.



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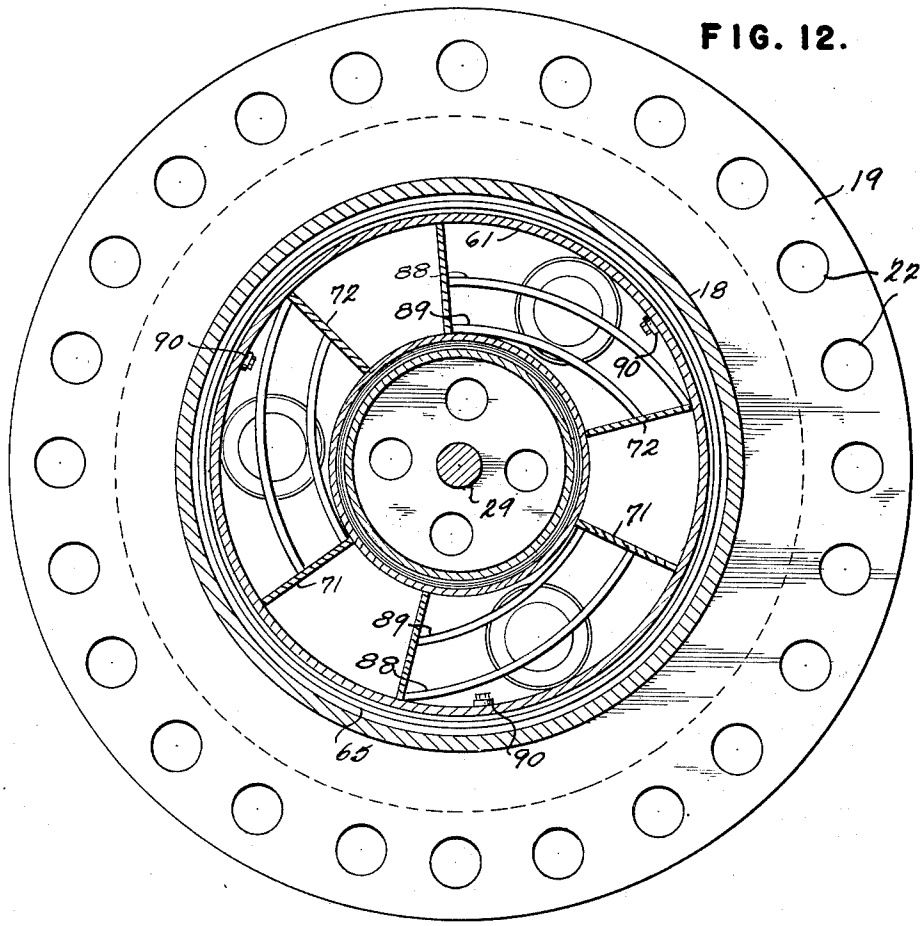
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METHOD OF CATALYTIC DESULFURIZATION OF GASES

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FIG. 12.



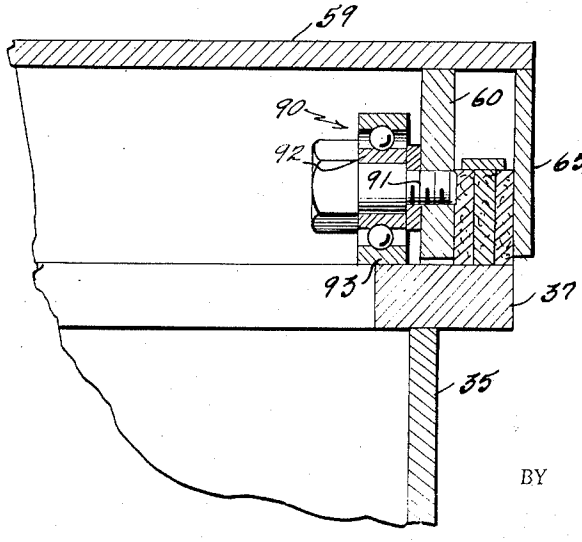


FIG. 13.

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METHOD OF CATALYTIC DESULFURIZATION OF GASES

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FIG. 14.

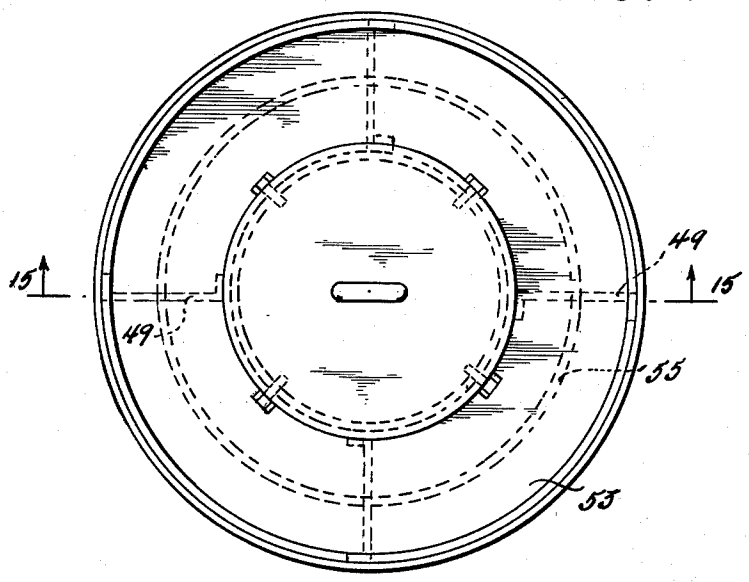
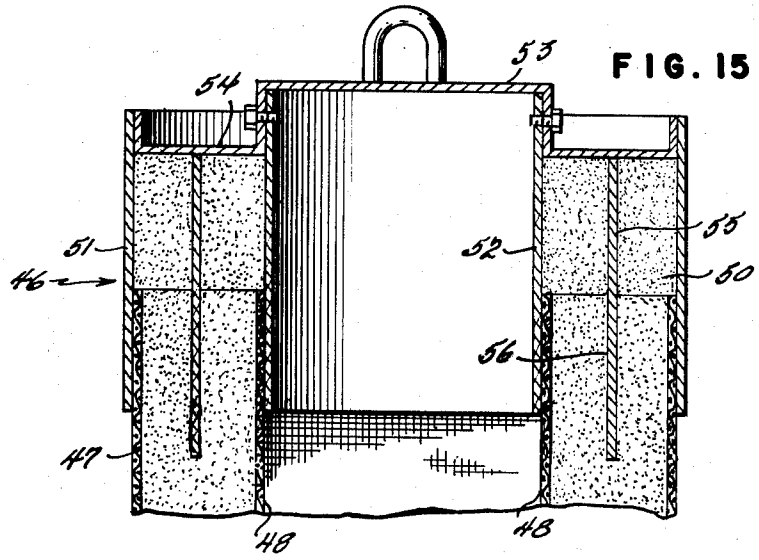


FIG. 15



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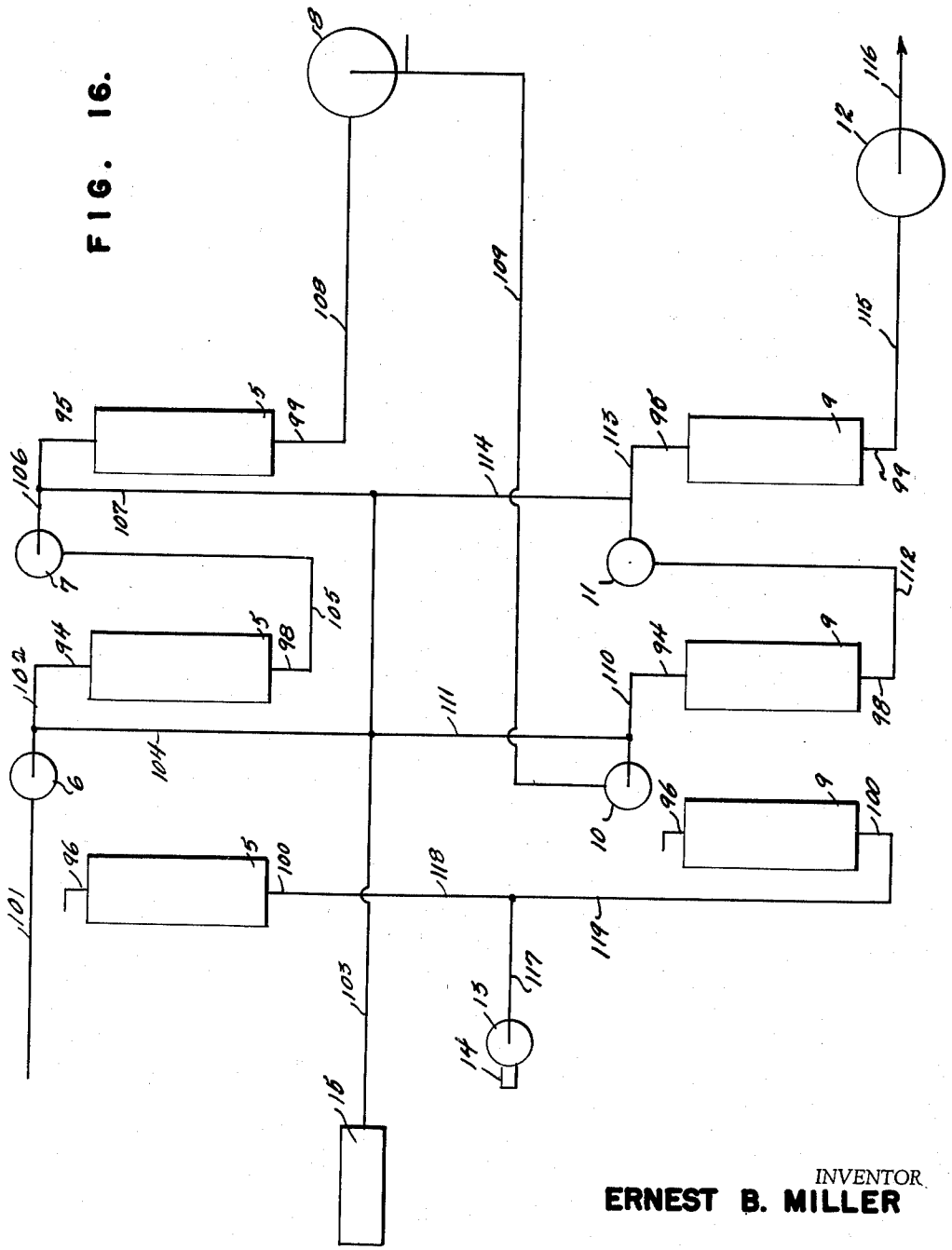
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March 3, 1953 E. B. MILLER 2,630,375

METHOD OF CATALYTIC DESULFURIZATION OF GASES

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FIG. 16.



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Patented Mar. 3, 1953

2,630,375

UNITED STATES PATENT OFFICE

2,630,375

METHOD OF CATALYTIC DESULFURIZATION OF GASES

Ernest B. Miller, Houston, Tex., assignor to Jefferson Lake Sulphur Company, New Orleans, La., a corporation of New Jersey

Application March 12, 1951, Serial No. 215,181

10 Claims. (Cl. 23—226)

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This invention relates to the recovery of sulphur from gases containing sulphur compounds and has more particular reference to a novel method of recovering elemental sulphur in liquid form from gases containing hydrogen sulphide.

The object of the present invention is to provide a novel method of recovering elemental sulphur from gas containing hydrogen sulphide by catalytically desulphurizing the gas to obtain liquid sulphur.

Another object of the invention is to provide a novel method of recovering elemental sulphur from gases containing H_2S in which a catalyst is used which is able to effect a highly efficient conversion of H_2S to H_2O and sulphur and in which the catalyst, after it has become spent, due to chemical reduction of the catalytic agent or the deposition of carbon or other contaminants from the gas being treated, may be fully restored to its initial efficiency by reactivation with hot air.

Another object of the invention is to provide a novel method of recovering elemental sulphur from gases containing H_2S, as characterized above, wherein the gas to be treated is brought into contact with the catalyst in at least two successive reaction zones, in which sulphur vapor is formed, prior to its passage to a scrubbing or condensing zone in which the sulphur vapor is condensed to form liquid sulphur.

Another object of the invention is to provide a novel method of recovering elemental sulphur from gases containing H_2S, as characterized above, wherein a portion of the liquid sulphur being recovered is returned to a sulphur burner, where it is burned to supply the oxidant required to oxidize the H_2S in the gas being processed.

A further object of the invention is to provide a novel method of recovering elemental sulphur from bases containing H_2S, as characterized above, wherein the temperature rise in each reaction zone is controlled by controlling the supply of oxidation gas to such zones.

Other objects and advantages of the invention will appear in the specification when considered in connection with the accompanying drawings, wherein:

Fig. 1 is a plan view, with parts omitted, showing one form of apparatus and the arrangement thereof for carrying out the method of the present invention;

Fig. 2 is a side elevational view of the apparatus shown in Fig. 1, and showing the second converter and its appurtenances and the second scrubbing tower and its appurtenances;

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Fig. 3 is a side elevational view of the first converter and its appurtenances, shown in Fig. 1;

Fig. 4 is a side elevational view of the first scrubbing tower and its appurtenances, shown in Fig. 1;

Fig. 5 is a plan view of a converter;

Fig. 6 is a side elevation, partly in section, of the converter driving mechanism;

Fig. 7 is a vertical sectional view of the converter, taken on the line 7—7 of Fig. 5, but omitting the driving mechanism;

Fig. 8 is a horizontal sectional view of the converter, taken on the line 8—8 of Fig. 7, but drawn to a smaller scale;

Fig. 9 is an enlarged vertical sectional view of the seal shown in the right hand side of the upper manifold of Fig. 7;

Fig. 10 is a plan view of the seal shown in Fig. 9;

Fig. 11 is a vertical sectional view taken on the line 11—11 of Fig. 10;

Fig. 12 is a horizontal sectional view taken on the line 12—12 of Fig. 7;

Fig. 13 is a partial vertical sectional view of a manifold showing the details of a roller;

Fig. 14 is a plan view of a tubular catalyst containing unit;

Fig. 15 is a vertical sectional view, with parts broken away, taken on the line 15—15 of Fig. 14, and

Fig. 16 is a diagrammatic view showing the flow of the gas to be desulphurized; the flow of the oxidation gas; and the flow of the regenerating medium through the system.

In general, the invention comprises a method of catalytic desulphurization of gases containing hydrogen sulphide, and the recovery of elemental sulphur therefrom in liquid form.

For the purpose of illustration, the invention will be described in connection with the catalytic desulphurization of sour natural gas and the recovering of elemental sulphur therefrom in liquid form.

Referring now to the drawings, there is shown, in Figs. 1 to 4 inclusive, one embodiment of apparatus and the arrangement thereof for carrying out the method of this invention. The apparatus shown includes a first three-stage rotary catalytic converter or reactor 5, two stages of which are used as reaction chambers or zones, in which the sour gas is brought into intimate contact with the catalyst, and the other stage is used as an activation chamber or zone in which the catalyst is regenerated; a first pre-heater 6 for heating the sour gas prior to its passage through the first reaction stage of the converter

5; a heat exchanger 7 for heating or cooling the sour gas between the first and second reaction stages of the converter 5; a first scrubbing tower 8 for separating the liquid sulphur from the treated gas; a second three-stage rotary catalytic converter 9, two stages of which are used as reaction chambers or zones in which the sour gas is brought into intimate contact with the catalyst and the other stage is used as an activation chamber or zone in which the catalyst is regenerated; a second pre-heater 10 for heating the sour gas after its passage through the first scrubbing tower and prior to its passage through the first reaction stage of the second converter 9; a second heat exchanger 11 for heating or cooling the sour gas between the first and second reaction stages of the second converter 9; a second scrubbing tower 12 for separating the liquid sulphur from the gaseous admixture after its passage through the second converter; a heater 13 for heating the regenerating medium, preferably air; a fan or blower 14 for supplying air under pressure to the heater 13; a sulphur burner 15, in which a mixture of liquid sulphur and air is burned to produce a gas of combustion having as high an SO_2 content as possible, preferably 19–20%; and a waste heat boiler 16 which utilizes the heat of the gases of combustion in the sulphur burner to generate steam for running auxiliary machinery and other purposes. The sulphur burner 15 and the waste heat boiler 16 may be of any suitable, usual type. The burner 15 is shown as having a liquid sulphur supply pipe 17 connected to supply liquid sulphur from the scrubbing towers, and an air supply pipe 18 connected to the hot air blower 14 for supplying air under pressure to the burner.

The two converters or reactors 5 and 9 are preferably supported in an upright raised position by suitable framework, indicated generally at 19 and 20, respectively.

The rotary converters 5 and 9 are identical in construction and are generally similar to the converter shown in my copending application, Ser. No. 779,824, filed October 14, 1947, for Method of and Apparatus for Desulphurization of Gases, now U. S. Letters Patent No. 2,561,990, issued July 24, 1951. The details of the converter are shown in Figs. 5 to 15, inclusive. As there shown, each of the converters comprises a pressure vessel 21, upper and lower manifolds 22, 23 fixedly mounted within the vessel; a compartmentized annular drum 24 rotatably mounted within the pressure vessel between and in communication with the manifolds; and suitable driving mechanism for rotating the annular drum.

The pressure vessel 21 is preferably formed in two parts, an upper flanged shell or can 25 and a lower flanged shell 26 suitably joined together, as by bolting, to form a gas-tight joint.

The annular drum 24 is fixedly attached as by means of plates 27, 28, to a central vertical shaft 29 suitably journaled in bearings carried by the upper and lower shell members 25, 26. The mechanism for rotating the annular drum is supported on a platform 30 mounted on the upper shell 25 and includes a shaft 31 connected to the upper end of the shaft 29 by a coupling 32 (see Fig. 6). The shaft 31 is driven by suitable reduction gearing mounted in a housing 33, the reduction gearing being belt-driven by a motor 34 (see Fig. 5).

The rotatable annular drum 24 comprises two spaced concentric cylinders 35, 36, which form the side walls; two spaced annular plates 37, 38, each secured to the top of the cylinders 35, 36, respectively, form the top of the drum, the space between the annular plates 37, 38 forming an annular opening 39 in the top of the drum; two spaced concentric annular plates 40, 41, each secured to the bottom of the cylinders 35, 36, respectively, form the bottom of the drum, the space between the annular plates 40, 41 forming an annular opening 42 in the bottom of the drum (see Fig. 7).

The rotatable annular drum is divided into a plurality of compartments 43 by radial partitions or diaphragms 44 (see Fig. 8). In each of the radial compartments 43, near the bottom thereof, there is provided a plate 45 attached to the walls of the compartment, as by welding, to form a gas-tight joint (see Fig. 7). Each plate 45 forms a support for one or more tubular catalyst containers 46. In the particular embodiment shown, only one such container is shown mounted in each compartment.

The catalyst containers 46 are identical in construction and, as shown in Figs. 14 and 15, each comprises two concentric tubular wire screens 47, 48 held in spaced relation by a plurality of longitudinal radial fins 49, with the annular space between the screens closed at the bottom. The mesh of the screen is such as to retain a granular catalyst material 50 in the annular space between the screens. Although the invention is not limited thereto, it is preferred to employ a catalyst wherein granular silica gel or a substance having substantially the same structure is the carrier for the active material. Oxide of copper, nickel, and manganese may be employed as the active material. However, iron oxide is preferred. In connection with the foregoing, it may be pointed out that the annular beds of catalyst material are so thin that they permit velocities of about from 10 to 25 feet per minute through the beds at 60°–70° F. and atmospheric pressure.

Each of the containers 46 is closed at its top by means of concentric hoops 51, 52 mounted on the concentric screens 47, 48 and a cover plate 53 detachably connected to the inner hoop 52, as by screw bolts, and having a depending annular trough-shaped flange 54 fitting between the hoops 51, 52. A depending annular fin 55 is secured to the flange 54 and projects downwardly between and below the hoops 51, 52, and fits in slots 56 formed in the upper ends of the radial fins 48, all as shown in Fig. 15. The construction is such that, as the catalyst settles down, leaving a space between the top portion of the wire screens devoid of catalysts, the fin 55 will prevent gas from passing through the space devoid of catalyst. Each container 46 is detachably mounted on a nozzle 57 projecting upwardly from an opening 58 formed in the plate 45, as clearly shown in Fig. 7. The nozzle 57 is secured in the opening 58, as by welding, to form a gas-tight joint.

The top and bottom manifolds 22, 23 are mounted on the top and bottom of the annular drum 24, in communication with the annular openings 39, 42 formed in the top and bottom of the drum. The manifolds are identical in construction and each is formed in the shape of an annular trough having an annular top (or bottom) 59 and annular side walls 60, 61 (see Fig. 9).

A plurality of compression springs 62, mounted on brackets 63 suitably secured to the inner walls of the vessel 21, yieldably press the top and bottom manifolds against the top and bottom, respectively, of the annular drum (see Fig. 7). The

top and bottom manifolds are held stationary relative to the rotation of the drum by means hereinafter to be described, and, to prevent the escape of gas between the rotating drum and the manifolds, sealing ring gaskets 64 are placed at the junction of the side walls of the manifolds and the drum. The sealing ring gaskets 64 are held in tight sealing engagement with the top and bottom of the drum by means of annular hoops 65 which encircle the gaskets and hold them against the side walls of the manifold. The upper (or lower) ends of the hoops 65 are secured to the top (or bottom) plate of the manifold, as by welding. The ring gaskets are retained between the hoops 65 and the side walls 60, 61 of the manifolds by means of a plurality of circumferentially spaced threaded bolts 66, which engage the ring gaskets and the lower portions of the hoops and side walls. The ring gaskets 64 are yieldably held in engagement with the top and bottom of the drum 24 by means of a plurality of compression springs 67 mounted on stud bolts 68 secured to the top (or bottom) of the manifolds and engaging annular plates or members 69 mounted on the top (or bottom) of the ring gaskets, all as clearly shown in Fig. 9.

At three circumferentially spaced points in the top and bottom manifolds, there are located seals which, by reason of the sliding contact of the radial partitions 44 against the under surface of the bottoms of the seals, divide the manifolds and drum into three sectors, each sector gas-tight with respect to the adjacent sectors. The seals are identical in construction and the details thereof are best shown in Figs. 9, 10 and 11. Each seal includes a bottom or sealing plate 70 mounted within the manifold between spaced radial partition walls 71, 72. The bottom plate 70 is yieldably urged against the top (or bottom) of the drum and rests on the concentric plates 37, 38 which form the top of the drum (or plates 40, 41 which form the bottom of the drum), as shown in Fig. 9. The side edges of the plate 70 are bifurcated, as shown at 73, 74, for the reception of gasket strips 75, 76, which are yieldably pressed outwardly against the partition walls 71, 72 of the seal by leaf springs 77, 78, as shown in Figs. 10 and 11.

The means for yieldably pressing the bottom plate 70 of the seal between the top (or bottom) of the drum comprise a plurality of compression springs 79 mounted on projections 80, formed on the upper surface of the plate 70. The springs 79 engage the top (or bottom) of the seal and are held in position by bolts 81 projecting through the top (or bottom) of the seal and coiled springs and threaded into the projections 80 formed on the plate 70.

Each radial partition or diaphragm 44 has a portion of its top and bottom edges extending upwardly (or downwardly) between the edges of the openings in the top and bottom of the drum. A gasket 82 is secured on these portions and extends above (or below) their top (or bottom) edges and engages the under face of the bottom plate 70 of the seal (see Fig. 11).

Plates 83 are secured to the tops and bottoms of the partitions and are held spaced therefrom by a spacer strip 84, the plates and spacer strip being secured to the partitions by bolts 85. The gaskets 82 are confined between the partitions and the plates 83, as by means of bolts 86, and are pressed upwardly (or downwardly) against the under surface of the bottom plates 70 of the seals by means of leaf springs 87, all as shown in Fig. 11.

In order to prevent the gaskets 82 from being unduly pressed upwardly (or downwardly) when the gaskets are not engaging the bottoms of the seals, means are provided for spanning the reaches of the manifolds between the seals. These means comprise spaced pairs of curved plates 88, 89 which extend between and are secured to the partition walls of the seal, as shown in Fig. 12. The bottom surfaces of the plates 88, 89 are in the same horizontal plane as the bottom surfaces of the bottom plates 70 of the seals, so that, as the gaskets 82 move out of engagement with the bottom plate of the seal, they immediately engage the plates 88, 89.

A plurality of rollers 90 are mounted within the top and bottom manifolds. These rollers are circumferentially spaced within the manifolds and are adapted to engage the annular plates 37, 40 which form parts of the top and bottom, respectively, of the rotatable drum. These rollers are adapted to prevent frictional surface engagement between the side walls of the manifolds and the top and bottom of the drum. These rollers are identical in construction and mounting and each comprises a threaded stud bolt 91 screwed into the outer side wall 60 of the manifold; a ball race 92 fixedly mounted on the bolt; and a wheel 93 mounted on the ball race, all as shown in Fig. 13.

Three flanged pipes or conduits 94, 95, 96 having threaded ends, project through the cap of the vessel 21 and have their threaded ends secured to the top plate of the top manifold by means of lock nuts 97 which form gas-tight joints (see Fig. 7). The pipes are welded to the cap and hold the top manifold stationary relative to the rotation of the drum. The three pipes are circumferentially spaced with respect to the top manifold and each is secured to and communicates with the manifold at a point located between the seals.

Three additional flanged pipes 98, 99, 100, having threaded ends, project through the bottom of the vessel 21 and have their threaded ends secured to the bottom plate of the bottom manifold by means of lock nuts 97 which form gas-tight joints. These pipes are welded to the bottom of the vessel 21 and hold the bottom manifold stationary relative to the rotation of the drum. These pipes are circumferentially spaced with respect to the bottom manifold and each is secured to and communicates with the manifold at a point located between the seals. The width of the seals with respect to the radial compartments 43 containing the catalyzing units is such that at all times at least one of the partitions or diaphragms 44 is engaging the bottom plate 70 of the seal in gas-tight engagement.

From the foregoing, it will readily be seen that by the engagement of the radial partitions with the seals, the manifolds and drum are divided into three gas-tight chambers or sectors, called, for convenience, the first reaction stage, the second reaction stage, and the activation stage.

The drum carrying the tubular catalyst containers is rotated counter-clockwise, as viewed in Fig. 5, and, as it rotates, the tubular catalyst containers are successively moved through the three stages in the following order: the second reaction stage, the first reaction stage, and the activation stage.

The flow of the gas to be treated, the flow of the oxidant gas, and the flow of the regeneration medium are shown schematically in Fig. 16.

The sour gas to be treated is delivered, under

suitable pressure from a source of supply (not shown), to the first pre-heater 6, by means of a pipe line 101. The gas is heated in the pre-heater to an optimum pre-reaction temperature and then passes through a pipe line 102 to the first converter 5 and enters the top manifold of the first reaction stage through pipe 94.

The oxidant gas, preferably SO_2, for the first reaction stage of the converter 5, is delivered under suitable pressure, from the sulphur burner 15 through a supply pipe line 103 and a branch valved pipe line 104 connected to the pipe line 102. The mixture of sour gas and SO_2 passes downwardly from the upper manifold through the opening in the top of the drum into the various compartments of the drum containing the tubular catalyst containers, as are at that time contained within the sector forming the first reaction stage. The gaseous mixture passes through the pervious layer of catalyst material into the hollow interiors of the tubular containers; thence, downwardly through the opening in the plate 45 into the bottom of the drum, and through the opening therein into the bottom manifold. When the mixture of sour gas and SO_2 comes into intimate contact with the catalyst an exothermic reaction takes place in which elemental sulphur is liberated in vapor form, together with steam, in accordance with the following equation:

$$2H_2S+SO_2=2H_2O+3S$$

From the bottom manifold, the treated gaseous admixture passes through pipe 98 and pipe line 105 into the first heat exchanger 7, where its temperature, which has risen in the reactor, is adjusted to an optimum pre-reaction temperature. From the first heat exchanger 7, the gaseous admixture passes through pipe lines 106 and 95 into the top manifold of the second reaction stage of the converter 5.

The oxidant gas, preferably SO_2, for the second reaction stage of the converter 5, is delivered under suitable pressure through a branch valved pipe line 107 connecting the supply line 103 and the pipe line 106. The gaseous admixture moves downwardly through the second reaction stage of the converter 5, in a manner similar to its downward movement through the first reaction stage and during its passage further reaction takes place, liberating more elemental sulphur vapor and steam.

From the second reaction stage of the first converter 5, the treated gaseous admixture passes through pipe 99 and pipe line 108 into the bottom of the first scrubbing tower 8. The treated gas mixture including the sulphur vapor and steam formed by the reactions in the converter, rise in the scrubbing tower against a downward flow of molten sulphur, which condenses the sulphur vapor into molten sulphur, which collects in the bottom of the tower.

From the top of the first scrubbing tower 8, the partially desulphurized gas passes through pipe line 109 into the second pre-heater 10, where its temperature is adjusted to an optimum pre-reaction temperature. From the second pre-heater, the gas passes through pipe line 110 and pipe 94 of the second converter 9 into the top manifold of the first reaction stage of the second converter 9.

The oxidant gas, preferably SO_2, for the first reaction stage of the converter 9 is delivered under suitable pressure through a branch valved pipe line 111 connecting the supply line 103 and the pipe line 110. The gaseous admixture moves downwardly through the first reaction stage of the second converter in a manner similar to its downward movement through the first reaction stage of the first converter and during its passage further reaction takes place, liberating more elemental sulphur vapor and steam.

From the first reaction stage of the second converter the treated gaseous admixture passes through pipe 98 and pipe line 112 into the second heat exchanger 11, where its temperature, which has risen in the reactor, is adjusted to an optimum pre-reaction temperature.

From the second heat exchanger 11, the treated gaseous admixture passages through pipe line 113 and pipe 95 of the second converter 9 into the top manifold of the second reaction stage of the second converter 9.

The oxidant gas, preferably SO_2, for the second reaction stage of the second converter is delivered, under suitable pressure, through a branch valved pipe line 114 connecting the supply pipe line 103 and the pipe line 113. The gaseous admixture moves downwardly through the second reaction stage of the second converter in a manner similar to its downward movement through the first reaction stage of the first converter and during its passage further reaction takes place and the remaining elemental sulphur is liberated in vapor form.

From the second reaction stage of the second converter, the treated gaseous admixture passes through pipe 99 of the second converter 9 and pipe line 115 into the bottom of the second scrubbing tower 12. The treated gaseous admixture, together with the steam and sulphur vapor formed by the reactions in the second converter, rises in the second scrubbing tower against a downward flow of molten sulphur, which condenses the sulphur vapor into molten sulphur which collects in the bottom of the scrubbing tower. From the top of the second scrubbing tower, the now sweet gas passes through a pipe line 116 to its point of use (not shown).

As schematically shown in Fig. 16, air for use as the regenerating medium is forced into the hot air heater 13 by means of the fan or blower 14. The air is heated to a temperature of about 1000° F. in the heater and, from the heater, passes through supply pipe line 117 and branch pipe line 118 to the first converter 5 and enters the bottom manifold of the activation stage through pipe 100. From the bottom manifold, the hot air passes through the opening in the bottom of the drum into the bottoms of the various compartments of the drum as are at that time contained within the reactor forming the reactivation stage; thence, upwardly through the openings in the plates 45, and up into the hollow interiors of the tubular catalyst containers, through the pervious layers of catalyst material into the compartments of the drum; thence, upwardly through the opening in the top of the drum into the top manifold. As the hot air passes through the catalyst material, any impurities in the form of tars or carbonaceous matter are burned off and the catalyst material is reactivated. From the top manifold of the activation stage, the hot air is exhausted to the atmosphere through pipe 96.

Hot air for use as the regenerating medium for the reactivation stage of the second converter passes from the air supply pipe 117 and branch pipe line 119 to the second converter and enters the bottom manifold of the reactivation stage

through pipe 100 of the second converter. The hot air passes upward through the reactivation stage of the second converter in a manner similar to its upward passage through the reactivation stage of the first converter and, during its passage, regenerates the catalyst material therein.

While the foregoing description of the method has not been concerned with the utilization of pressurized equipment, this is not a limitation. Some pressure will be required on the system to overcome the back pressure due to friction. Pressure above this limitation is not necessary to successful operation. On the other hand, by allowance for the differential pressure, a reasonable increase in the over-all operating pressures will permit handling larger gas volumes in an apparatus of fixed size. The particular apparatus shown is designed to handle gases at high pressures; although it is contemplated that the method herein described can be practiced by utilizing pressures of from about 5 lbs. to 10 lbs. per square inch.

The present method is contrived to recover a major portion of the contained sulphur in the sour gas in a multi-stage oxidation process without raising the reaction temperature in any stage above about 830° F. by adjusting the temperature of the admixture of the gas to be treated and the sulphur liberating gas, illustratively SO_2, to an optimum pre-reaction temperature in the range of from about 375° F. to about 600° F. prior to the entry of the admixture into each of the stages or reaction zones and controlling the amount of SO_2 admitted into each stage or reaction zone. However, it is contemplated that substantial portions of the contained sulphur may be removed in the respective stages when employing pre-reaction temperatures up to about 800° F., while controlling the reaction temperatures in the stages so that they do not exceed about 1000° F. in any stage. It will be understood, however, that at higher reaction temperatures than above 830° F., the efficiency of the conversion will be reduced. Therefore, it is highly preferable to practice the method at relatively low reaction temperatures.

As a specific example, assume that the raw gas to be treated contains 125 lbs. of H_2S per 4300 cubic feet at a pressure of 10 lbs. gauge and at 100° F. Then, about 30% of the initial H_2S content of the gas can be converted into sulphur vapor in the first oxidation stage of the first group of reactors by pre-heating the gas to about 500° F. and mixing SO_2 with the pre-heated gas at the rate of about 30 lbs. of SO_2 per minute prior to the entry of the gas into the reactor in which the first oxidation stage occurs. The temperature in the reactor will rise to from about 720° F. to about 750° F.

About 30% of the initial H_2S content of the gas can be converted into sulphur vapor in the second oxidation stage of the first group of reactors by cooling the gaseous admixture delivered from the first oxidation stage to about 500° F., then mixing SO_2 with the cooled gaseous mixture at the rate of about 30 lbs. of SO_2 per minute prior to its entry into the reactor in which the second oxidation stage occurs. The temperature in the reactor will rise to from about 680° F. to about 710° F.

About 30% of the initial total H_2S content of the gas can be converted into sulphur vapor in the first oxidation stage of the second group of reactors by pre-heating the gas after its passage through the first scrubbing tower to about 500° F. and mixing SO_2 with the pre-heated gas at the rate of about 30 lbs. of SO_2 per minute prior to its entry into the reactor in the second group of reactors in which the first oxidation stage occurs. The temperature in the reactor will rise to from about 670° F. to about 700° F. The remaining 10% of the initial total H_2S content of the gas can be converted into sulphur vapor in the second oxidation stage of the second group of reactors by cooling the gaseous admixture delivered from the first oxidation stage to about 500° F., then mixing SO_2 with the cooled gaseous admixture at the rate of about 11 lbs. of SO_2 per minute prior to its entry into the reactor of the second group of reactors in which the second oxidation stage occurs. The temperature in the reactor will rise to from about 530° F. to about 560° F.

Obviously, with a raw gas having a much lesser H_2S content, it will be possible to remove all of the H_2S in the first group of reactors without raising the temperature in either the first or second oxidation stages above a permissible reaction temperature of from about 747° F. to about 830° F.

It is always advisable, however, to do as much oxidation as possible in the first stage, consistent with the above mentioned reaction temperatures, so that it has been found advisable, when treating a gas containing about 15% H_2S, to supply oxidant gases to the various stages so as to accomplish about 55% conversion in the first stage, about 25% in the second stage, about 12% in the third stage, and 8% in the last stage. Since, in the earlier stages, the catalytic conversion can not be completed, the reaction in these stages can be assisted by the presence of an excess of oxidant gas. Thus, to accomplish the illustrative reactions above, it has been found convenient to supply about 75% of the total oxidant gas in the first stage and the remaining 25% in the second stage. The excess not used up in either of these stages is carried with the gas to be treated, and is available as needed in the last two stages.

In connection with the foregoing, it may be pointed out that, as the boiling point of sulphur is 832° F., a pure sulphur vapor would condense to liquid if cooled below that temperature. In the present method, as above described, there is no concentrated sulphur at any point. In fact, the maximum concentration is about 2%, at which concentration the sulphur vapor will remain in vapor form at the pressures and temperatures employed in the process.

While it is not a necessary feature of the invention, it may be pointed out that, if the liberated sulphur content of the treated gas becomes too high between the first and second oxidation stages of either group of reactors, all or part of it may be removed. This is readily accomplished, for example, by suitable adjustment of the heat exchangers 7 and 11.

The scrubbing towers 8 and 12 may be of any suitable usual type. In the particular embodiment illustrated, molten sulphur is withdrawn from the bottom of the scrubbing tower 8 through pipe line 120 by pump 121 and delivered to a sulphur cooler 122 through a pipe line 123. From the sulphur cooler 122, the molten sulphur passes through pipe line 124 to the top of the scrubbing tower 8. The molten sulphur cascades downwardly through the scrubbing tower and is brought into intimate contact with the counter-current stream of gas, steam

and elemental sulphur vapor rising through the scrubbing tower and condenses the sulphur vapor into molten sulphur which collects in the bottom of the tower. In like manner, molten sulphur is withdrawn from the bottom of the scrubbing tower 12 through a pipe line 125 by a pump 126 and delivered to a sulphur cooler 127 through a pipe line 128. From the sulphur cooler 127, the molten sulphur passes through a pipe line 129 to the top of the scrubbing tower 12 and cascades downwardly through the tower in counter-current to the upward flow of gas, steam and sulphur vapor therein and condenses the sulphur vapor, which collects in the bottom of the tower. A portion of the molten sulphur being recirculated through the two scrubbing towers, is withdrawn through a pipe line 130, which is connected to a cross pipe line 131 connecting the discharge pipe lines 124 and 128 from the sulphur coolers 122 and 127, and delivered in its molten state to a sulphur cooling and flaking machine (not shown). Pipe line 17, through which liquid sulphur is supplied to the sulphur burner 15, is shown as being connected to pipe line 130.

By mounting the annular drum and the manifolds of the converter within pressure vessels, the method may be carried out with high pressure gases and, too, the equalization of pressure within the drums, manifolds and vessels, permits the drums and manifolds to be made of lighter weight material, which adds considerably to the efficient and economical operation of the converters. This equalization is accomplished by means of a small opening 132 formed in that portion of the pipe line 94 within the vessels 21. It is also considered desirable to provide each of the six pipes 94, 95, 96, 98, 99 and 100 of each of the converters with expansion joints 133, located a short distance from their points of connection to the manifolds. This may be necessary, due to the unequal temperatures of the gases and media as they pass through the various sectors, thereby resulting in unequal expansion of the parts of the drums and manifolds.

While two three stage rotary converters have been shown, obviously, one converter may be used, if desired, in which case, the treated gas, after passing through the first scrubbing tower 8, would be sent to its further point of use by means of a valved by pass conduit 134 connected to valved conduit 109.

From the foregoing, it readily will be seen that there has been provided a novel method of recovering elemental sulphur in liquid form from gases containing H_2S, which provides for carrying out the reaction process in at least two oxidation stages with control of the temperature rise in each stage, thereby permitting the reaction process to be effected at temperatures below about 800° F., with resulting increase in efficiency of the process, use of less expensive apparatus, and permitting the use of a catalyst carrier such as silica gel.

While the invention has been described in connection with the desulphurization of sour gas, obviously, it is also applicable to the desulphurization of other types of sulphur-containing gases and vapors which are subject to the action of a suitable catalyst.

Obviously, the invention is not restricted to the particular embodiment thereof herein shown and described. Moreover, it is not indispensable that all of the features of the invention be used conjointly, since they may be employed advantageously in various combinations and subcombinations.

What is claimed is:

1. In the recovery of elemental sulphur in liquid form from gases containing H_2S involving the contact of a catalyst with the gas containing H_2S to form sulphur vapor and the subsequent treatment of the catalyst in an oxidizing atmosphere to regenerate the catalyst for further contact with the gases containing H_2S and the subsequent condensation of the sulphur vapor to form liquid sulphur, the improvement which comprises rotating a series of separated thin beds of catalyst directly and in succession and substantially continuously relative to and through a series of reaction zones and a regenerating zone; continuously directing the flow of the gas to be treated in succession and in series through said reaction zones; subjecting the gas to be treated to heat exchange to bring its temperature to an optimum reaction temperature and mixing a predetermined amount of oxidant with it prior to its passage through each of the reaction zones to convert a portion of the H_2S in each of said zones into sulphur vapor and steam; continuously withdrawing the treated gas from the last one of the reaction zones and directing its flow through a condensing zone and there condensing the sulphur vapor to form liquid sulphur; and continuously directing the flow of a hot oxidizing medium through the regenerating zone to reactivate the catalyst therein.

2. The method, as set forth in claim 1, wherein the direction of the series flow of the treated gas is opposite to the direction of rotation of the catalyst beds, whereby the treated gas will always make its last passage through freshly activated catalyst beds.

3. The method, as set forth in claim 1, including rotating a second series of separated thin beds of catalyst directly and in succession and substantially continuously relative to and through a second series of reaction zones and a regenerating zone; continuously withdrawing the treated gas from said condensing zone and directing the flow of the withdrawn gas in succession and in series through said second succession of reaction zones; subjecting the withdrawn gas to heat exchange to bring its temperature to an optimum reaction temperature and mixing a predetermined amount of oxidant gas with it prior to its passage through each of said second series of reaction zones to convert a portion of the H_2S in each of said second series of reaction zones into sulphur vapor and steam; continuously withdrawing the treated gas from the last one of the reaction zones of said second succession of reaction zones and directing its flow through a second condensing zone and there condensing the sulphur vapor to form liquid sulphur; and continuously directing the flow of a hot oxidizing medium through the regenerating zone of said second succession of zones to reactivate the catalyst therein.

4. The method, as set forth in claim 1, including the step of burning a mixture of sulphur and air to make SO_2 and supplying the SO_2 for use as the oxidant.

5. In the recovery of elemental sulphur in liquid form from gases containing H_2S involving the contact of a catalyst with the gas containing H_2S to form sulphur vapor, the subsequent treatment of the catalyst in an oxidizing atmosphere to regenerate the catalyst for further contact with

the gas containing H_2S and the subsequent condensation of the sulphur vapor to form liquid sulphur, the improvement which comprises rotating a series of beds of catalyst directly and in succession and substantially continuously relative to and through a series of reaction zones and a regenerating zone; continuously directing the flow of the gas to be treated in succession and in series through said reaction zones; subjecting the gas to be treated to heat exchange to bring its temperature to an optimum reaction temperature and mixing with the gas to be treated an amount of SO_2 sufficient to convert a portion of the H_2S into sulphur vapor and steam, without raising the temperature of reaction in any of the zones above about 800° F. prior to its passage through each of the reaction zones; continuously withdrawing the treated gas from the last one of said reaction zones and directing its flow through a condensing zone and there condensing the sulphur vapor to form liquid sulphur; and continuously directing the flow of a hot oxidizing medium through the regenerating zone to reactivate the catalyst therein.

6. The method, as set forth in claim 5, including rotating a second series of separated thin beds of catalyst directly and in succession and substantially continuously relative to and through a second series of reaction zones and a regenerating zone; continuously withdrawing the treated gas from said condensing zone and directing the flow of the withdrawn gas in succession and in series through said second succession of reaction zones; subjecting the gas to be treated to heat exchange to bring its temperature to an optimum reaction temperature and mixing with the gas to be treated an amount of SO_2 sufficient to convert a portion of the H_2S into sulphur vapor and steam without raising the temperature of reaction in any of the zones above 800° F. prior to its passage through each of the reaction zones; continuously withdrawing the treated gas from the last one of the reaction zones of said second succession of reaction zones and directing its flow through a second condensing zone and there condensing the sulphur vapor to form liquid sulphur; and continuously directing the flow of a hot oxidizing medium through the regenerating zone of said second succession of zones to reactivate the catalyst therein.

7. The method, as set forth in claim 5, including the step of withdrawing a portion of the liquid sulphur from the condensing zone and burning it to supply the SO_2.

8. In the recovery of elemental sulphur in liquid form from gases containing H_2S involving the contact of a catalyst with the gas containing H_2S to form sulphur vapor and the subsequent treatment of the catalyst in an oxidizing atmosphere to regenerate the catalyst for further contact with the gases containing H_2S and the subsequent condensation of the sulphur vapor to form liquid sulphur, the improvement which comprises rotating a series of separated thin beds of catalyst directly and in succession and substantially continuously relative to and through a reaction zone and a regenerating zone; mixing a predetermined amount of the gas to be treated with a predetermined proportion of sulphur-liberating gas and continuously directing a flow of the mixed gases at an optimum pre-reaction temperature through said reaction zone to convert the H_2S in said reaction zone into sulphur vapor and steam; continuously withdrawing the treated gas mixture from said reaction zone and directing its flow through a condensing zone and there condensing the sulphur vapor to form liquid sulphur; and continuously directing the flow of a hot oxidizing medium through the regenerating zone to reactivate the catalyst therein.

9. The method, as set forth in claim 8, including the step of burning a mixture of sulphur and air to make SO_2 and supplying the SO_2 for use as the oxidant.

10. The method as set forth in claim 9, in which the sulphur is obtained by recycling a portion of the recovered liquid sulphur.

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