June 22, 1965 H. PULS 3,190,812

DEVICE FOR THE CONTINUOUS PRODUCTION OF HARD PITCH

Filed Jan. 17, 1961 2 Sheets-Sheet 1

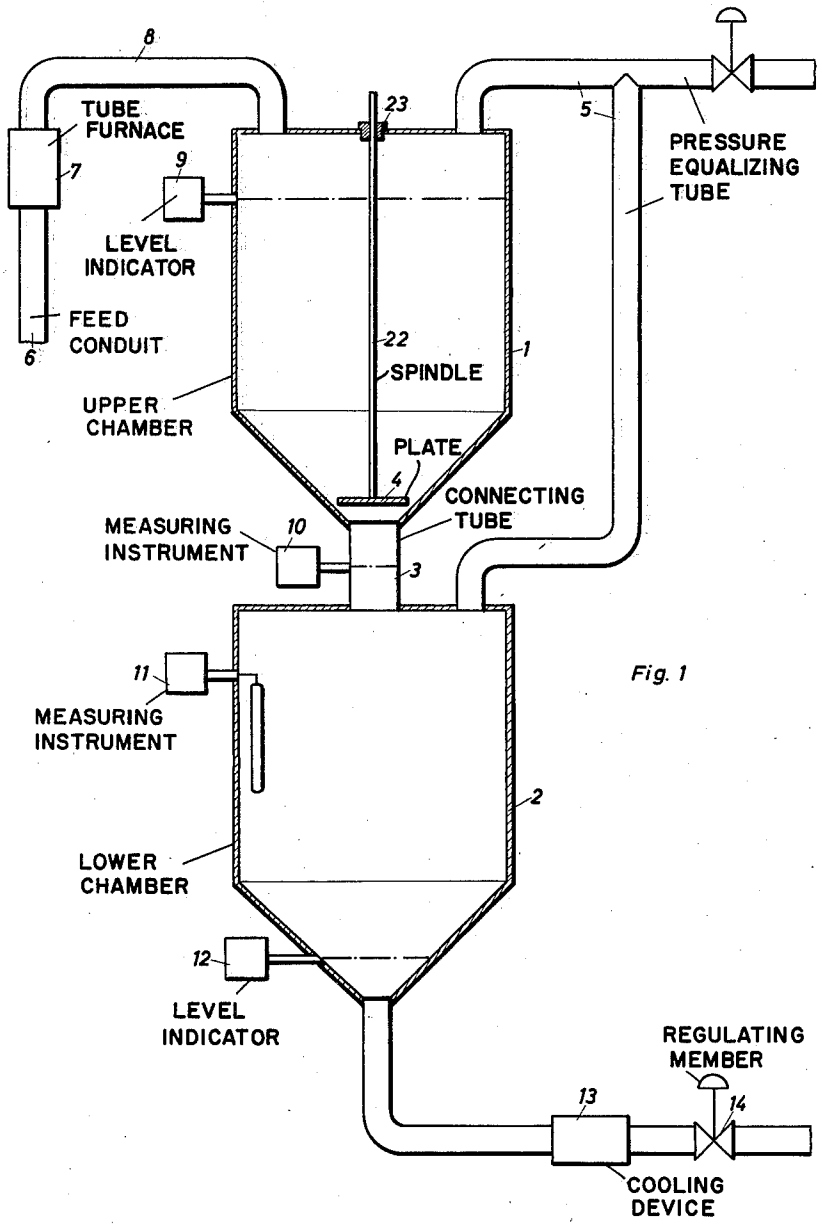


Fig. 1

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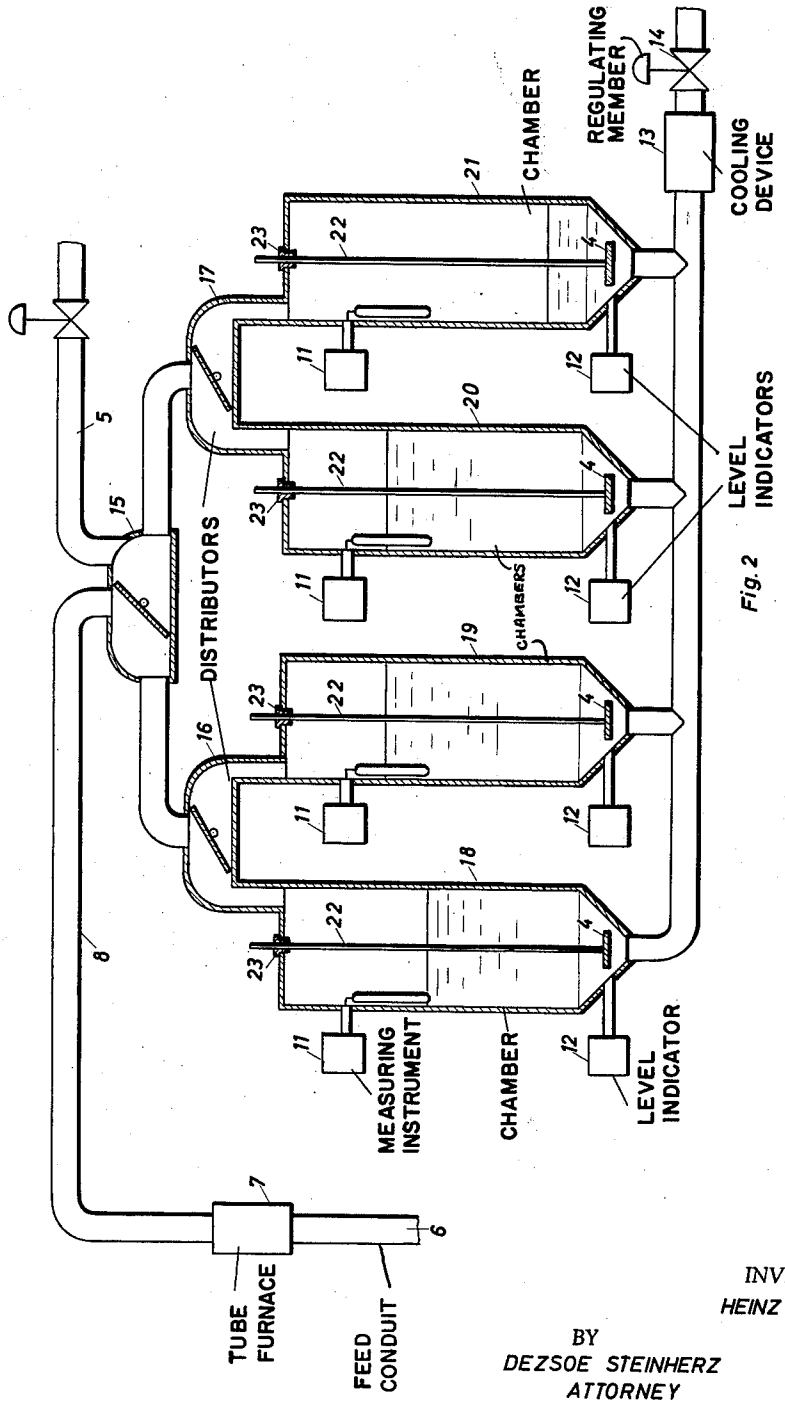


Fig. 2

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DEVICE FOR THE CONTINUOUS PRODUCTION OF HARD PITCH

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Claims priority, application Germany, Jan. 22, 1960, R 27,160
2 Claims. (Cl. 196—107)

This invention relates to the production of hard pitch and it has particular relation to the production of hard pitch by thermal treatment of softer pitches.

According to the process of prior application Serial No. 690,392, filed in the name of Julius Geller et al. on October 15, 1957 for "Process for the Treatment of Tars and Tar Products" (now Patent No. 2,985,577) coal tars, distillation products of coal tars and mixtures of these materials, are converted into hard pitch by passing said materials through a tube furnace under heating and under a pressure of 3–10 atmospheres to a discharge temperature in the range of 450°–525° C., discharging the materials from the tube furnace substantially without the loss of heat and pressure, into a thermally insulated reaction vessel, in which the materials undergo further treatment under a pressure in the range of 3–10 atmospheres, without supply of heat to said vessel, and discharging the materials from said vessel when their desired conversion into hard pitch is attained.

If this process is carried out continuously, in some cases the use of a pressure vessel through which the material treated continuously flows is adequate, if the amount of material treated in the vessel is kept constant and the amount discharged is regulated in dependence on the degree of filling. The feed supplied mixes then extensively with the reacting material in the vessel and the discharged material is a mixture consisting of pitch particles which were subjected to reaction for different periods of time.

If it would be possible to follow up the individual pitch particles after their entrance into the reaction vessel, it could be observed that some particles pass immediately to the discharge opening and leave the pressure vessel without having participated in the reaction, while other particles remain in the vessel and do not reach the discharge opening. The reaction time of all pitch particles lies between these two borderline cases. The average time of stay, which is calculated from the relation of the contents of the vessel to the amount supplied and the amount discharged, permits a reliable statement on the character of the end product. However, there are relatively few particles which stay in the pressure vessel during a reaction period of equal duration. If the course of reaction is judged on the basis of determination of the coking residue in accordance with DIN DVM 3725, an almost uniform distribution of the coke contents of the individual particles in the end product results, starting from the initial value up to the end value, which would appear at the selected reaction temperature and infinite reaction time. The coking residue of the end product discharged reaches the same value which would result if all pitch particles were subjected to the reaction for the average period of stay.

However, difficulties may arise if mixing of the pitch entering the reaction vessel, with the contents of the reaction vessel, is incomplete. The occurs when those pitch particles which have been in the reaction vessel of a considerably longer period than the average period of stay, lose their flowing capacity and appear as colloids in the end product.

The present invention provides a process and device

which permit continuous thermal conversion also of those pitches which show thereby a tendency of being decomposed to heterogeneous end products.

According to this process, the supply of pitch to the reaction vessel and the discharge from said vessel takes place—as in the above mentioned prior process—continuously, but, according to the invention, within the vessel a periodic storage and passing on of the pitch to be forwarded through the vessel, is carried out. In order to attain this, the reaction vessel is subdivided into at least two chambers, which are arranged side by side, or preferably one above that other. While one of these chambers is filled with a stream of constant amount, the other chamber is emptied by the discharge of an equal amount of the stream, so that the sum of the content of the two chambers remains always constant. The filling, as well as the discharge take place during a period of time which corresponds to the average period of stay.

The process according to the present invention has the advantage that mixing of the freshly supplied product with the contents of the chamber takes place in one chamber only, while the other chamber, from which the material is discharged contains only highly reacted material, the total amount of which is discharged. The end product produced in this manner consists also of pitch elements reacted to varying extents; however, in comparison with the above described prior process the parts reacted for very short times are strongly reduced and the parts which have been reacted for very long reaction times and are undesired due to lack of flowing capacity have completely disappeared. Because in the mixing of pitch portions which have been reacted for a short time only, a homogeneous condition appears very quickly, the end product of the present process practically corresponds to the average 40% of the process, in which there is only a single reaction vessel through which the material constantly flows, while in the use of the subdivided reaction vessel according to this invention the 35% treated for a short time and the 25% reacted for a very long time, do not appear in the new mixture.

In the case of pitches, the treatment of which is unusually difficult and in cases, in which an extremely uniform end product must be maintained, the reaction vessel is subdivided into three, or still more, chambers. While one chamber is being filled and the other emptied, the charge in the other chambers is subject to undisturbed reaction, or, if the chambers are arranged one above the other, the charge is forwarded intermittently from one chamber to the other. In this case too the sum of the contents of all of the chambers is constant at any time. It is determined by the product of reaction time multiplied by the amount of the throughput. The time z for filling and discharging, respectively, of a chamber can be calculated by the number n of the chambers and the reaction time t, according to the equation

$$t=(n-1).z$$

The appended drawings illustrate some specific examples of and best modes for carrying out the invention, to which the invention is not limited.

In the drawings FIGURE 1 diagrammatically illustrates an apparatus comprising two reaction chambers arranged one upon another, while FIGURE 2 diagrammatically illustrates an apparatus comprising four chambers arranged side by side.

Example 1

In FIG. 1 the reaction vessel consists of two chambers 1 and 2 which are arranged one upon another and connected by a tube 3, the free cross-sectional area of which is large enough to allow passing of the pitch therethrough

in a very short time. The tube 3 can be closed by a plate 4, which is operated by a spindle which is guided by a stuffing box arranged in the upper horizontal wall of chamber 1. The closing by plate 4 need not be entirely tight in order to perform its job. The pressure in chambers 1 and 2 is equalized by the action of tube 5, through which the gases formed in the reaction escape. The starting material to be treated is introduced through conduit 6 into tube furnace 7 and is heated in the latter to reaction temperature. Through tubing 8 the product is introduced through the top into heat insulated chamber 1 and accumulates in chamber 1 at closed position of plate 4 until the level indicator 9 responds. Then plate 4 is lifted so that the material in chamber 1 passes to heat insulated chamber 2 as quickly as possible. If measuring instrument 10 indicates that tube 3 is not completely filled with liquid any more, plate 4 is lowered again to closing position.

From chamber 2, the material flows to cooling device 13, in which it is cooled to a temperature, at which practically no reaction takes place. A regulating member 14 keeps the discharge of material at a constant value.

Due to small differences between the amount of material fed and discharged, respectively, the total amount of material in the reaction vessel can change during a relatively long period of operation. In order to counteract this, the valve 14 can be opened or closed, slightly more or less at the time when according to the indication of leveling instrument 11, chamber 1 is completely emptied. Complete emptying of chamber 2 is prevented by level indicator 12. If the level of material in chamber 2 becomes lower than the range of indicator 12, the latter causes lifting of plate 4, although level indicator 9 has not responded yet.

Example 2

FIGURE 2 shows four thermally insulated chambers arranged side by side and preferably housed by a reaction vessel which is not shown in FIGURE 2. The means for feed and discharge are denoted in FIG. 2 by the same reference symbols as in FIGURE 1. In the position shown in FIG. 2, the distributors 15, 16 and 17 guide the pitch discharged from the tube furnace 7 into the last discharged chamber 18. At the same time, chamber 21 is discharged, while the material is reacted in chambers 19 and 20. When level indicator 12 shows that chamber 21 is empty, its discharge opening is closed and the discharge opening of chamber 20 is opened. The indication of measuring instrument 11 arranged on chamber 18 shows at that time whether the regulating valve 14 needs adjustment. After chamber 18 is filled, the distributors 15 and 17 are switched in order to bring about refilling of chamber 21.

In the above Example 1, the starting material is e.g. a briquetting coal tar pitch having an ash content of 0.15%, a softening point of 70° C. and a coking residue of 30%. This pitch is heated in the tube furnace to a discharge temperature of 490° C. under a superatmospheric pressure of 5 atmospheres and is then reacted in the insulated chambers shown in FIG. 1 for a total period of time of 25 minutes. The hard pitch thus obtained has an ash content of 0.16%, a softening point of 130° C. and a coke residue of 54%.

The same starting material can be treated in a substantially similar manner in the apparatus described in Example 2, under similar conditions and with similar results.

Instead of briquetting coal tar pitch, other pitches can be used, e.g. brown coal tar pitch, or blast-furnace tar pitch. Thereby, the temperature and pressure conditions, as well as the duration of treatment can be modified in dependence on the characteristics of the starting material and the desired properties of the end product.

It will be understood from the above that this invention is not limited to the steps, conditions, materials, devices and other details specifically described above and illustrated in the drawings and can be carried out with various modifications. For example, the starting material is treated by heating it in the tube furnace to a temperature in the range of 400 to 525° C. under a superatmospheric pressure in the range of 1 to 20 atmospheres, and the duration of the treatment in the reaction vessel may take 0.2 to 10 hours, depending on the temperature and pressure used and the desired characteristics of the hard pitch to be obtained. For example, a coal tar pitch having a softening point of 40 to 60° C. and a coke residue of 20 to 25% can be converted into a hard pitch having a softening point of 90 to 100° C. and a coke residue of 38 to 44% by heating said coal tar pitch in the tube furnace to 400 to 420° C. and treating it in the reaction vessel for 600 to 350 minutes. The steps, conditions, variations and features described in the above mentioned prior application Ser. No. 690,392, or in the corresponding British Patent No. 858,132, can be analogously applied in carrying out the present invention. The preferred starting material in carrying out this invention is a pitch, e.g. coal tar pitch, which yields 25 to 35% coke upon destructive distillation. The hard pitches obtained according to the present invention yield 50 to 60% coke upon destructive distillation. The ash content of hard pitches thus prepared only slightly exceeds the ash content of the starting material and the pitches prepared according to this invention are particularly suitable for the production of high grade electrode coke. For example, from a soft coal tar pitch having an ash content of 0.10 to 0.20%, a hard pitch having 0.11 to 0.21% ash content can be prepared according to the present invention. Operation of the device according to the present invention can be carried out substantially in the manner and under the conditions described in the examples of the above mentioned prior application Ser. No. 690,392 or of the corresponding British Patent No. 858,132. The products of this invention can be utilized for briquetting, as binding agents, adhesive masses, starting material for the manufacture of electrodes, for structural purposes, road building, manufacture of molded products and other purposes. The content of the coke residue is determined by the method of the German Standard DIN DVM 3725, which is quite similar to the standardized method described in the book: Abraham, Asphalts and Allied Substances, 5th edition, page 1132–34.

The reference symbols 4, 5, 6, 7, 8, 11, 12, 13, 14 have in FIG. 2 the same meaning as in FIG. 1. Reference symbol 22 denotes the spindle and 23 the stuffing box in both figures. Opening and closing of the discharge openings of chambers 18, 19, 20, 21 in FIG. 2 is brought about by displacement of plates 4 connected with the spindles 22. In a plant for commercial operation each chamber may have, for example, a capacity of receiving 100 to 25,000 kgs. of pitch. In such chambers the pitch can flow from an upper chamber to the chamber arranged below it, e.g. in 0.2 to 30 minutes.

The parts and percent mentioned herein are by weight if not otherwise stated.

What is claimed is:

1. A device for the continuous production of hard pitch from a softer pitch, pitch-containing materials and pitch-forming starting materials, essentially consisting of a tube furnace and a pressure vessel comprising at least two thermally insulated adjacent chambers which receive no heat supply and are arranged one above the other, adjacent chambers being connected by a tube, said chambers receiving from said tube furnace heated starting material flowing in downward direction to the highest chamber and from there to adjacent chambers arranged below said highest chamber means for alternately closing and opening said tube in order to effect alternate charging and discharging the chambers and means for equalizing the pressure in said chambers.

2. A device as claimed in claim 1, in which each chamber has a conical bottom and a cover part, and the cover part of at least one chamber is provided with a stuffing box, and the means for closing said tube consist of a plate operable by a spindle connected with the plate and guided by said stuffing box.

References Cited by the Examiner

UNITED STATES PATENTS

135,673	2/73	Tait et al.	196—111
1,965,074	7/34	Gray	196—107
1,980,839	11/34	Webre	196—107
2,006,188	6/35	Sydnor	196—111
2,032,546	3/36	McNeil et al.	196—111

2,120,296	6/38	Pier et al.	208—85
2,173,984	9/39	Shapleigh	23—212
2,326,186	8/43	Watson	208—50
2,573,149	10/51	Kassel	260—683.3
2,916,432	12/59	McNamara	208—2
2,937,138	5/60	Beuther	208—39
2,967,815	1/61	Dell et al.	208—213
2,983,665	5/61	Dell et al.	208—39
2,985,577	5/61	Geller et al.	208—6

OTHER REFERENCES

Sachanen: "Conversion of Petroleum," pp. 274 to 277, pub. by Reinhold Pub. Co., N.Y., 1940.

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