May 30, 1939. T. REICHERT 2,160,341

PROCESS FOR THE CARBONIZATION OF ORGANIC CELLULOSIC MATERIALS OF VEGETABLE ORIGIN

Filed Dec. 15, 1936 2 Sheets-Sheet 1

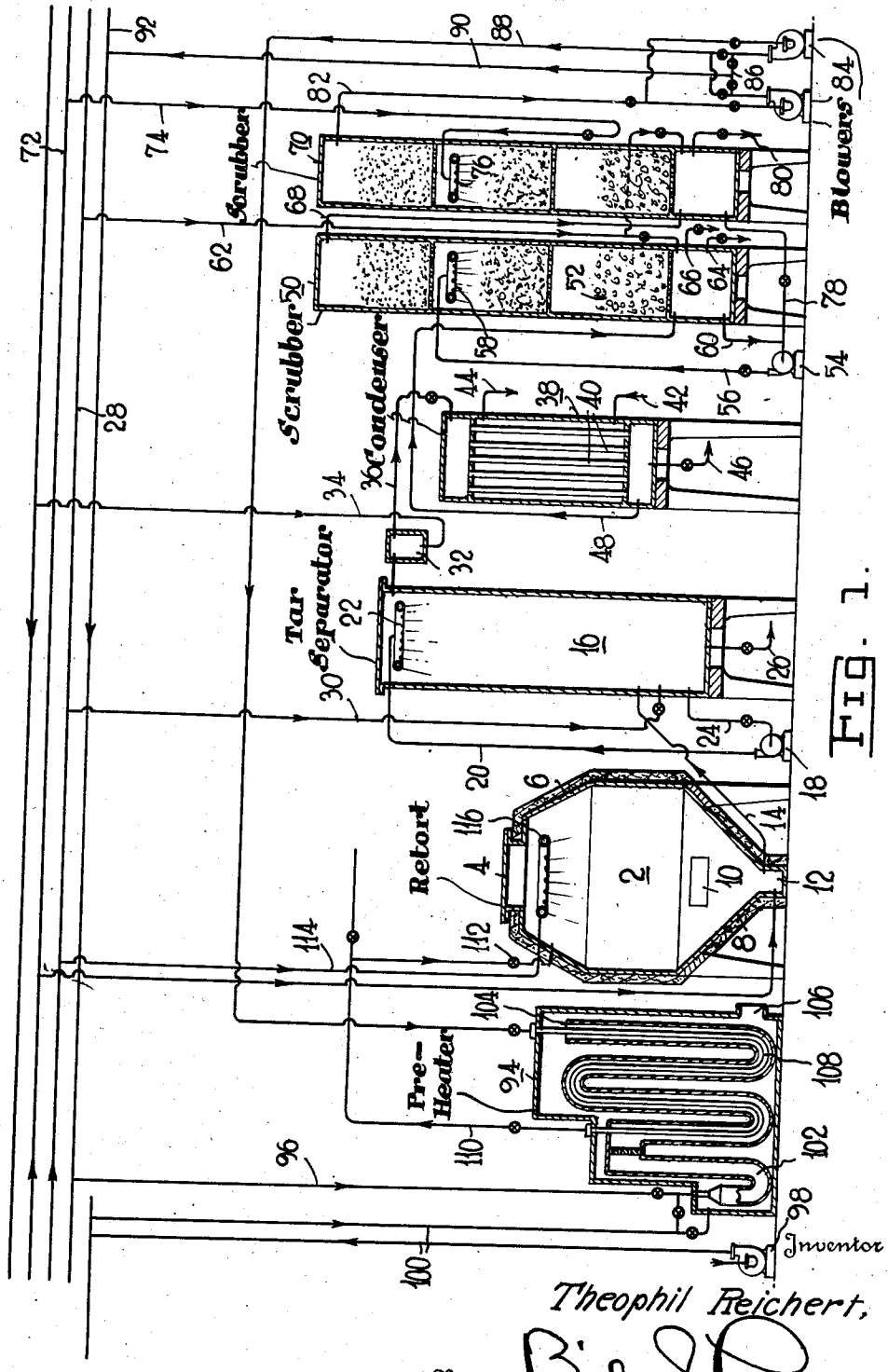

FIG. 1.

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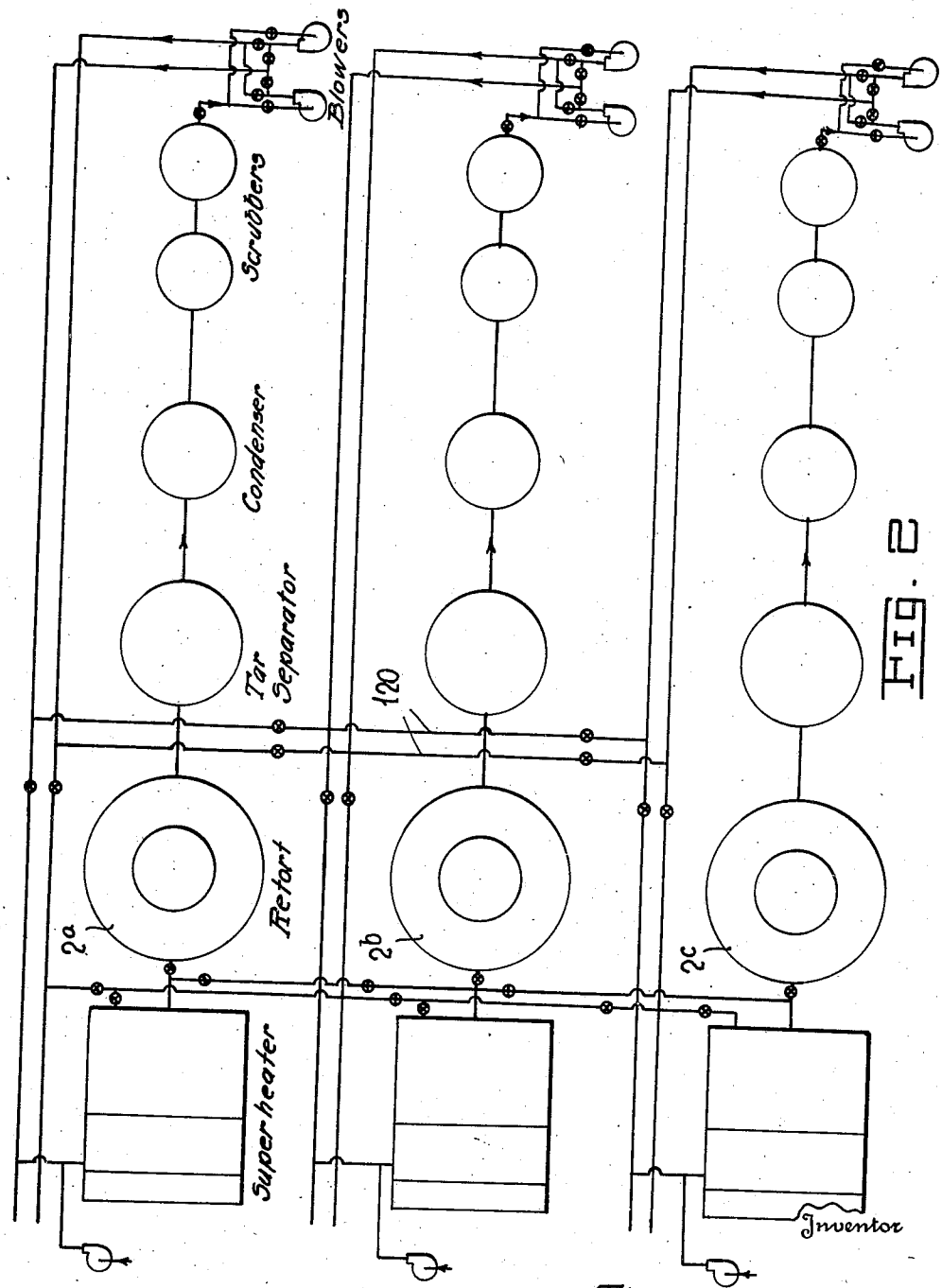

Fig. 2

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PROCESS FOR THE CARBONIZATION OF ORGANIC CELLULOSIC MATERIALS OF VEGETABLE ORIGIN

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Application December 15, 1936, Serial No. 116,034
In Germany December 24, 1932

5 Claims. (Cl. 202—16)

My invention relates to an improved process for the carbonization of organic cellulosic materials of vegetable origin, preferably of some form of wood, but also of similar materials such as nutshells, corn cobs, peet, lignin and the like.

This application is a continuation-in-part of my copending application, Serial Number 703,816, filed December 23, 1933.

It is a well known fact that in the dry distillation of such materials a decomposition reaction occurs at a certain temperature which is highly exothermic. At this point in the process a sudden and large rise in temperature occurs. This increased temperature causes undesirable decomposition of a large part of the valuable distillation products present in the wood, and thus decreases the yield of such products. Numerous efforts have been made to avoid this effect, but none of them have been successful.

I have found, surprisingly, that this decomposition of the distillation products can be avoided and higher yields obtained by a special manipulation of the carbonization process.

The principal feature of my discovery is that decomposition of this kind can be largely avoided if the material is heated from the top downwardly in the proper manner. In other words, the material is carbonized by hot gases which are introduced or formed at the top of the mass to be carbonized and drawn downward through the lower part of the mass in such a manner that the carbonization progresses gradually in zones from the top downwardly, so that simultaneous exothermic reaction of the entire contents of the mass is avoided.

According to a particular feature of the invention, the wood is carbonized by passing hot gases, which are properly tempered, from the top downwardly, in such a manner and at such a velocity that the zone-wise carbonization takes place beginning at the top. In other words, the temperature and velocity of the heating gases are so chosen that the upper layers are carbonized and then gradually this carbonization proceeds downwardly through the mass.

The process can be performed in many various types of retorts. Horizontal, vertical or inclined retorts may be used. In all these types of apparatus, heat losses should be avoided by insulating the retort.

The preferred manner of practicing the process consists in preheating a gas which does not contain free or uncombined oxygen and then drawing such heated gas downwardly through the material. The vaporous distillation products obtained from the wood pass downwardly with the heated gases and are drawn off at or near the bottom of the retort. I have found that in order to produce step-wise carbonization it is necessary that the rate at which the heating gases are passed through the wood should be between .1 and 3 cubic meters of gas per kilogram of wood per hour. In the preferred form of the invention from 1 to 2 cubic meters of gas are used per hour for each kilogram of wood.

Instead of using preheated gas, however, it is possible to burn a combustible gas just above or in the top of the heating chamber, while avoiding carefully any excess of oxygen, so that no free oxygen will be present in the gases. Instead of using gas as a fuel, coal-dust or oil might also be used. The use of gas is preferable as the regulation of heat is much easier with a gaseous fuel.

The process may also be carried out by heating the upper layers of the materials to be carbonized to an oxidation temperature in the presence of air or oxygen, thus producing hot gaseous products. These may be drawn through the material at a rate dependent on the supply of air to the upper layer, and preferably at such a rate as to provide oxygen-free gases in the proportion specified above. Another manner of carrying out the process is to heat the upper layers so as to produce gaseous distillation products which may be drawn down through the remainder of the material and themselves act as heating gases.

The method of operation above described produces a progressive heating from top to bottom through the mass and thus a progressive carbonization. Each portion of the mass beginning with the top layer is first dried and then carbonized and the gaseous products driven out. Inasmuch as the gaseous products from any part of the material are drawn downwardly immediately into a zone of lower temperature, they are subjected at once to this lower temperature and are subject to the carbonizing temperature only for a short time. In this manner any excessive decomposition of the products by long exposure to the carbonizing temperature is avoided. Upon entering the lower zone the heat of the reaction is absorbed by the relatively cool material still to be treated and thus the gases or gaseous products are cooled immediately. At the same time, the flow of gas is not so rapid as to cause carbonization of the lower layers before the upper layers have been carbonized.

According to a further feature of the inven-

tion, some of the gases issuing from the retort, preferably after the distillation products have been recovered from them, may be recirculated through the retort either alone or together with hot fresh gases. Furthermore, some of the evolved gases may be burned to supply heat to the circulating gases.

The process can be performed periodically, semi-periodically or continuously. With periodic operation the carbonization process can be performed fractionally.

One essential requirement of the process relates to the temperature and the rate of circulation of the heating gases. The rate of circulation of the gases should be sufficiently high so that there is no appreciable condensation of the condensible constituents driven off from the wood in the layers below the zone in which carbonization is taking place. In other words, while the gases must be immediately cooled to prevent further decomposition by the heat of carbonization, nevertheless they should not be cooled to such an extent as to produce condensation of the condensible constituents thereof. On the other hand, it is important that the rate should be sufficiently low so that the mass is heated gradually or in layers or zones to the carbonizing temperature. If the gas is introduced too rapidly, then the heat is carried down into the lower zones and all will be carbonized at substantially the same time. This would mean that any gases which are evolved in the upper layers would not be cooled and would be subjected to a considerable period of high temperature; and thus the distillation products would be likely to be decomposed, and the beneficial effects of the process would be lost.

The apparatus which is disclosed is particularly suitable for carrying out the process above generally described.

Further objects and advantages of the invention will appear more fully from the following description, particularly when taken in conjunction with the accompanying drawings which form a part thereof.

In the drawings:

Fig. 1 is a diagrammatic showing of an apparatus for carrying out the invention.

Fig. 2 is a diagram showing an apparatus in which three or more carbonizing retorts may be used.

The invention will be readily understood from a description of the accompanying drawings showing an apparatus in which this process may be carried out.

As shown in Fig. 1, the apparatus includes a carbonizing retort 2. This retort may be opened and closed at the top as at 4 to permit filling thereof. The upper wall section 6 and the lower wall section 8 are in the form of frustums of cones. An important feature of the invention is that the top wall section 6 must be at a greater angle of inclination or more nearly vertical than the lower wall 8, their angles being 57° and 45° respectively. As is shown by the drawings, the walls of the retort 2 are insulated to prevent the escape of heat therefrom. Materials such as the carbonized wood may be removed from the bottom of the retort through an opening 10 or in any other suitable fashion.

At the bottom of the retort is arranged a receiver 12 into which the gases pass. From the receiver 12 the gases pass through a line 14 to a tar separating column 16, for removing tar-like constituents. The gases enter near the bottom of this column. A pump 18 is provided which will force either tar or tar-oils through a pipe 20 to a spray 22 in the top of the column 16. The tar-oils are drawn off at the bottom through a pipe 24 and returned to the pump. An outlet pipe 26 may be provided at the bottom of the separating column through which excess tar may be removed.

It is also desirable at times to supply steam to mix with the gases passing through this column. This steam may be drawn from a line 28 and supplied through a pipe 30 to the bottom of the column 16.

From the top of the column 16 the gases may pass into a small chamber 32 filled with tar or any liquid to avoid gases from flowing back in the case that the pressure of gases before said chamber should decrease. From the chamber 32 the gases pass through a pipe 36 to the top of a condenser 38. The gases pass downwardly through the condenser through tubes 40, the space around these tubes being supplied with a cooling liquid through inlet pipe 42 and exit pipe 44. The bottom of the condenser is provided with an outlet 46 to permit the withdrawal of the condensate therefrom. Crude acetic acid and some crude wood alcohol are thus separated out.

From the bottom of the condenser a pipe 48 leads the remaining gases to the bottom of a scrubber 50. This scrubber is filled with porous material 52. The gases pass upwardly therethrough in counter-current relation with water which is pumped by a pump 54 through a pipe 56 to a spray 58 adjacent the top of the column. The water is returned through a pipe 60 to the pump. If desired, steam may also be introduced through the line 62 from the line 28 into the bottom of the scrubber, and the water and wood spirit which are washed out may be withdrawn separately through outlets 64 and 66.

From the scrubber the gases pass through a pipe 68 to the bottom of a second scrubber 70. This scrubber is also supplied with water from a line 72 through a pipe 74 to a spray 76 within the scrubber. The water from the bottom of the scrubber is withdrawn by a pipe 78 to the pump 54. Any excess water may also be taken off through the outlet 80. Wood spirit and the soluble volatile constituents are removed in these scrubbers.

A pipe 82 leads from the top of the scrubber 70 to conduct the gas to blowers 84. These blowers and a system of pipes and valves 86 permit the distribution of the gas to either of two pipes 88 and 90. The pipe 90 leads into a main 92, while the pipe 88 leads the gas to a preheating arrangement 94 now to be described.

Gas from the main 92 is supplied through a pipe 96 to the preheating arrangement 94. At the same time, an air pump 98 pumps air through pipes 100 to this device. In the preheater, the gases furnished by the pipe 96 and the air furnished by the pipe 100 are burned together in the space 102. This space is of a serpentine nature, and the gases flow therethrough as they are burned and come out at 104, from which they can pass out of the preheating device through a flue 106. The gases in pipe 88 on the other hand pass through the smaller pipes 108 inside the pipes 102, and are preheated by the gases being burned in the pipes 102. These preheated gases pass out through pipe 110 to a valve 112 arranged at the top of the carbonizing retort 2. Thus the gases produced in the retort are first cooled and washed so that all the condensible constituents

are removed and are then returned, after being preheated, and passed again through the retort in a downward direction to cause further carbonization of the wood.

If desired, steam may also be supplied from the main 28 through the pipe 114 to a spray 116 in the top of the retort 2. This is used to remove any residue of volatile products during or after the carbonization.

Obviously suitable valves are arranged in the various lines at the points shown in order to control the operation of the apparatus in the desired manner and to regulate the quantities of the various materials flowing through the pipes.

I will now describe the operation of the device shown and the manner in which it carries out the process which forms the subject matter of the invention.

The retort 2 is first filled with the material to be carbonized, as for instance the wood of deciduous trees, and hot gases from any suitable source, such as from another retort of the same type, are introduced through valve 112 into the top of the retort. These hot gases pass downwardly through the retort and out through the pipe 14. The gases first dry the wood and any water which is evaporated passes off with the gases through the outlet 14.

As soon as the wood is dried, the hot gases which pass downwardly through the wood begin to carbonize the same. These preheated gases drive out the distillation products from the wood and carry them downwardly. These gases pass out through the pipe 14 to the various parts of the apparatus described above. Such of the gaseous constituents as are of a tar-like nature are separated in the tar separating columns 16. The condensible constituents are condensed and separated in the column 38. Other volatile or soluble constituents are washed out in the scrubbers 50 and 70. The remaining portion of the gases, which have a high fuel value, is pumped by the blowers 84 through the pipe system 86 to various points of use.

A certain portion of the gases are pumped through the pipe 88 and then through the preheater pipes 108 to the top of the retort. Some of the other gases are pumped into the line 92 and then through pipe 96 to the outer preheater pipes 102 where they are burned to heat the gases flowing through the inner pipe. The gases to be supplied to the retort are thus preheated by the burning of some of the products of the heating of the wood.

The gases introduced at the top of the retort 2 preferably have a temperature of between 500° C. and 600° C. The circulation of the gases is carried out at such a rate by means of the blowers and valves that the quantity of gas flowing through the retort is between .1 and 3 cubic meters of gas per kilogram of wood in each hour. Preferably, the quantity is between 1 and 1.8 cubic meters. By this procedure the gases coming out at the bottom of the retort are at a relatively low temperature. During the greater part of the carbonization, the temperature of the escaping gaseous products is only about 30° C., although when the carbonizing zone approaches the bottom of the retort it may rise as high as 200° C.

By varying the amount of the recirculating gas within the limits specified, it is possible to obtain varying results. For example, the amount of tar produced can be varied so that more valuable products are obtained. Inasmuch as the circulating gases contain wood alcohol, an increased formation of esters takes place. Furthermore, the carbon content of the charcoal may be regulated so that it is possible to obtain a product containing 90% carbon.

The procedure above described has numerous advantages. A recirculation of the gases makes it possible to use a great proportion of the heat developed, since this heat is used for the heating of further gases. In this way the external heating required by the process is considerably reduced. Furthermore, the yield of acetic acid is increased, it being possible to obtain as much as 42 kilograms from the same amount of wood from which previous procedures had only yielded 32 kilograms.

It will thus be seen that the essential feature of the invention resides in passing the vaporous products from the wood with the hot gases downward immediately at a suitable rate of flow into a cooler zone where they are not subject to further decomposition. In this manner, the heat developed by the exothermic reaction is used in a sense to preheat but not to carbonize the lower layers of material.

In the form of the invention shown in Fig. 2, I have shown how three carbonizing retorts may advantageously be used simultaneously. During the time that one of these retorts 2*a* is being operated to carbonize wood, the contents of the second retort 2*b* may be prepared by treatment with hot gases to obtain a charcoal with increased carbon content, while the third retort 2*c* is being charged. The gases from one retort may be used in another through pipes 120, either as drying or heating gases or for any other purpose.

While I have described herein some embodiments of my invention, I wish it to be understood that I do not intend to limit myself thereby except within the scope of the appended claims.

I claim:

1. A process for the dry distillation of organic cellulosic distillable material of vegetable origin, in a retort heated internally only, which comprises supplying a hot gas at the top of a bed of such material of substantial depth within said retort to distill the material destructively while avoiding decomposition of the volatile substances evolved from the material by heating, passing said gas while still hot downwardly through the material in the retort at a rate of between 0.1 and 3 cubic meters per hour per kilogram of the material to be carbonized, the gas being fed at such a velocity and in such a quantity that the carbonization of the material proceeds gradually downwardly, that the temperature difference throughout the mass is maintained low, and that condensation of the evolved volatile substances in the lower part of the material is substantially prevented, and recovering distillation products from such gas.

2. A process for the dry distillation of organic cellulosic distillable material of vegetable origin, in a retort heated internally only, which comprises supplying a hot gas at the top of a bed of such material of substantial depth within said retort to distill the material destructively while avoiding decomposition of the volatile substances evolved from the material by heating, passing said gas while still hot downwardly through the material in the retort at a rate of between 1 and 1.8 cubic meters per hour per kilogram of the material to be carbonized, the gas being fed at such a velocity and in such a quantity that the carbonization of the material proceeds

gradually downwardly, that the temperature difference throughout the mass is maintained low, and that condensation of the evolved volatile substances in the lower part of the material is substantially prevented, and recovering distillation products from such gas.

3. A process for the dry distillation of organic cellulosic distillable material of vegetable origin, in a retort heated internally only, which comprises supplying a hot gas at the top of a bed of such material of substantial depth within said retort to distill the material destructively while avoiding decomposition of the volatile substances evolved from the material by heating, passing said gas while still hot downwardly through the material in the retort at a rate of between 0.1 and 3 cubic meters per hour per kilogram of the material to be carbonized, the gas being fed at such a velocity and in such a quantity that the carbonization of the material proceeds gradually downwardly, that the temperature difference throughout the mass is maintained low, and that condensation of the evolved volatile substances in the lower part of the material is substantially prevented, recovering distillation products from such gas, and returning at least a part of the gas from which distillation products have been recovered to the top of the retort.

4. A process for the dry distillation of organic cellulosic distillable material of vegetable origin, in a retort heated internally only, which comprises supplying a hot gas to the top of a bed of such material of substantial depth within said retort to distill the material destructively while avoiding decomposition of the volatile substances evolved from the material by heating, passing said gas while still hot downwardly through the material in the retort at a rate of between 0.1 and 3 cubic meters per hour per kilogram of the material to be carbonized, the gas being fed at such a velocity and in such a quantity that the carbonization of the material proceeds gradually downwardly, that the temperature difference throughout the mass is maintained low, and that condensation of the evolved volatile substances in the lower part of the material is substantially prevented, removing condensible constituents from such gas, heating at least a part of the gas from which the condensible constituents have been removed, and returning such heated gas to the top of the retort.

5. A process for the dry distillation of organic cellulosic distillable material of vegetable origin, in a retort heated internally only, which comprises supplying a hot gas to the top of a bed of such material of substantial depth within said retort to distill the material destructively while avoiding decomposition of the volatile substances evolved from the material by heating, passing said gas while still hot downwardly through the material in the retort at a rate of between 0.1 and 3 cubic meters per hour per kilogram of the material to be carbonized, the gas being fed at such a velocity and in such a quantity that the carbonization of the material proceeds gradually downwardly, that the temperature difference throughout the mass is maintained low, and that condensation of the evolved volatile substances in the lower part of the material is substantially prevented, removing condensible constituents from such gas, burning a portion of the gas from which condensible constituents have been removed and utilizing the heat thereof to preheat a second portion of the gas, and returning such heated portion to the top of the retort.

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