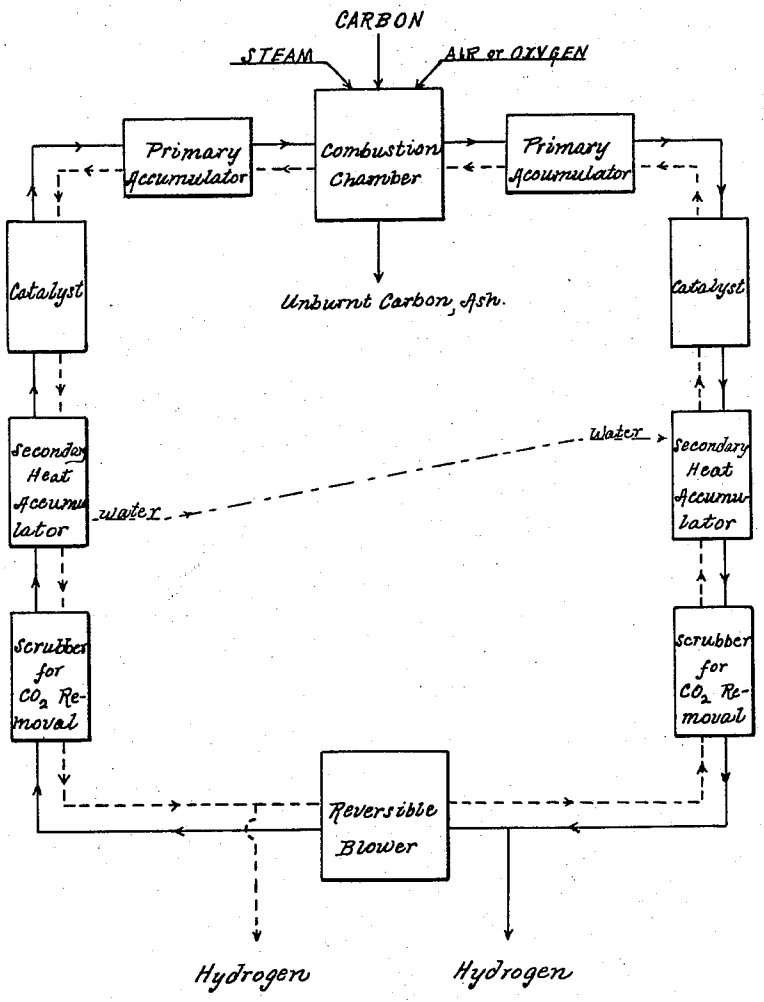
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METHOD OF MAKING HYDROGEN AND APPARATUS THEREFOR

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METHOD OF MAKING HYDROGEN AND APPARATUS THEREFOR

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This is a continuation-in-part of a former application bearing the same name and the Serial Number 231,967, filed June 16, 1951, and now abandoned. In said former application I took a very broad view of the raw material that could be used while in the instant application I stress certain features particularly evident when the raw material employed be charcoal. In this manner I am able to describe and illustrate to better advantage the salient divergence between my own technique and that now in use in the industry of making hydrogen.

In the actual chemistry involved there is, of course, no such divergence. The reactions between carbon and steam, whether said carbon be present as solid fuel, or as carbon monoxide, or even as a hydrocarbon, is too well known in the practice of today to require any elucidation. It is assumed that the reader is already familiar with all such matters. As already stated in said former application, and now repeated, I avail myself of the reaction whereby carbon is oxidized by the oxygen resident in steam, the over-all effect being the formation of carbon dioxide and the liberation of the resident hydrogen in said steam as hydrogen gas. The technique employed is best elucidated by considering the drawing submitted herewith.

It is, of course, a well known fact that at temperatures below 1200° F. the principal reaction between carbon and steam is that represented by the following equation: $2H_2O+C=CO_2+2H_2$—35,000 B.t.u., but if carbon be employed in the form of coal of any type then residual coke reacts very slowly and thus virtually forces the abandonment of this desirable reaction for the high temperature conversion represented by the conventional practice, namely, $H_2O+C=CO+H_2$—52,850 B.t.u. Obviously, therefore, the use of charcoal as the acepted form of carbon, owing to its much greater reactivity, permits the use of the low temperature reaction.

In the drawing, therefore, let the combustion chamber be filled with charcoal and the temperature maintained at approximately 1200° F. by commingling sufficient oxygen with the entering steam to supply the deficiency in the endothermic nature of said reaction, said oxygen forming CO_2 rather than CO. The products of said reaction then undergo the following treatment. By means of the reversible blower, shown at the bottom of the drawing, a stream of gas is sent through the reaction chamber, alternately in opposite direction, and the products of reaction commingle with said reversing gas flow. On either side of the combustion chamber I have shown a "heat accumulator" in which heat is either abstracted from or imparted to the gaseous fluid every time the direction of said flow is reversed. Following the "primary heat accumulator" I have indicated a pair of "catalyst" chambers and immediately thereafter another pair of "secondary heat accumulators" which in turn either abstract from or impart heat to the reversing gas stream. Finally I have shown a pair of "scrubbers" in which the carbon dioxide formed elsewhere is removed

by contact with an alkaline medium, preferably a caustic alkali solution at boiling temperatures, to avoid condensation of steam.

Obviously, there are cheaper ways of removing CO_2 than the one indicated, for the sake of simplicity. Whatever means be used is immaterial so far as the operation of my process is concerned. If the secondary accumulators be large enough then virtually all steam can be condensed, removed as water, and then re-evaporated in the corresponding secondary accumulator. Actual removal of CO_2 is best of all effected by the use of ammonia, an old application, in the event that the hydrogen be used in the manufacture of ammonia, but that demands cooling to room temperature as an imperative condition.

Returning now to the drawing. I have used the expedient of representing the reversing flow by two parallel lines and directional arrows, one line being solid and the other a broken or "dotted" line. Starting then at the blower and following the dotted line in the direction of the arrow the first device, the scrubber, is encountered. The composition of the gas from the blower is virtually hydrogen with a little CO, virtually all the CO_2 produced having been abstracted in the scrubber traversed by the gas before entering the blower. This right hand scrubber, therefore, has virtually no function until reversal of flow, so the gas passes through substantially unchanged. The amount of steam commingled with said hydrogen depends, of course, on the temperature to which the gas was cooled before entering the blower, and hence, is optional. To avoid confusion I have shown a "dot-dash" line connecting the two secondary heat accumulators to indicate that water condensed in one is re-evaporated in the other one. The function of the right hand, secondary heat accumulator is thus simply that of heating the traversing gas to the temperature suited to the catalyst which is selected from any conventional practice where it is desired to react steam and carbon monoxide for the production of hydrogen and carbon dioxide, an exothermic reaction which adds further heat to the gas stream.

Still following the dotted line from said catalyst chamber to the right hand "primary heat accumulator" the gas flow is now essentially steam and hydrogen with almost non-existent CO, the amount of this gas present at the blower having been converted to CO_2 in the highly efficient catalyst contact due to previous removal of CO_2. In the accumulator stored heat from the previous reversal is now again absorbed raising the gas stream to the reaction temperature desired in the combustion chamber, i. e., below 1200° F. On traversing this chamber, an amount of additional hydrogen and some carbon dioxide is added to the gas stream, the little CO_2 already present passing virtually unchanged through the incandescent fuel at this low temperature. The sensible heat in said gas is then stored in the left hand primary accumulator making the temperature that desired for the catalyst. A further increment is stored in the secondary left-hand accumulator and the gas is thus cooled for the scrubber and final removal of CO_2. In passing through the left hand catalyst any CO present is largely, though not completely, oxidized to CO_2 with attendant evolution of hydrogen from the steam commingled therewith. Thus is produced the gas "consisting essentially of hydrogen" which passes through the blower and reaches the point of origin of this description.

On reversal of flow, following now the solid line in the direction of the arrow it is obvious that the same set of circumstances will be in effect and will produce the same result. I have indicated the hydrogen thus produced removed at the pressure side of the blower, a

solid and a dotted line being used to designate said removal. The salient feature of my process is thus seen to be a reversing flow of steam and hydrogen picking up an increment of carbon dioxide from the charcoal and parting with same at the scrubber, the one regulator of the entire operation being the amount of oxygen introduced to satisfy the heat requirement of the reactions.

Considering this use of charcoal as the carbon source for my process I have thus given a concise and complete version of an illustration in which every item of said process is specifically pointed out. By following this technique the end result will be easily ascertained. Those specific items that may be deemed missing are comon knowledge to anyone familiar with the theme. It is obvious that the combustion chamber must be kept filled with charcoal, and it is equally obvious that steam must always be in excess. In conventional work it is often the practice to have five times the amount of steam present that is actually reacting as such an excess favors the reaction. The only variants are in the time for reversal and in amount of oxygen added. This again is obvious. Timing in reversal is a function of the size of the accumulators. When the temperature at any one point exceeds an arbitrary value, empirically determined, then it is time to reverse. Similarly, if the temperature in the combustion chamber exceeds 1200° F. it is time to reduce the ovygen. If the temperature drops too low it will at once show up by decreased quantity of the hydrogen produced. Again, as the amount of CO_2 produced has an adverse effect I deem it advisable not to permit too large a concentration before the scrubber removes it. As a limit I place a gas consisting of three times the amount of hydrogen, by weight, as carbon dioxide, this immediately before the scrubber removes it en toto. To decrease said amount obviously only means to increase the speed of the blower, thus re-cycling more pure hydrogen. Had I omitted all reference to said items it would have made no difference. Any person competent to manage such a plant will certainly know enough to supply any apparent deficit in information of which I may be guilty deeming all such material as technical knowledge possessed by any operator.

With a clear knowledge of my process as thus applied to charcoal it will be easy to understand modifications. Thus if the use of coal renders a high temperature mandatory, then the CO_2 will be formed but slightly in the combustion chamber and the gas will consist essentially of hydrogen and CO, but when this passes the catalyst conversion to CO_2 takes place only to an increased degree. In other words the locus for CO_2 formation passes from the combustion chamber to the catalyst chamber. Every thing else remains unchanged. Similarly, if the temperature of the combustion chamber be held at above 2200° F., at which temperature CH_4 decomposes into hydrogen and carbon, then natural gas can substitute for any other type of carbon, the liberated "lamp black" being in turn decomposed by steam in accord with the reactions previously cited. All such minor modifications I consider as within the scope of this disclosure.

As another modification the substitution of air for oxygen becomes a factor. In the event that ammonia be the ultimate objective then a mixture of almost theoretical composition, the 3:1 ratio, is readily obtained. In this case final adjustment is made by the admission of a very small amount of oxygen commingled with said air, such admixture being determined by gas analysis and empirical procedure.

Apparatus for this entire operation is conventional as to design though original in operation and results. It is evident that a combustion chamber must be of refractory construction, and this same observation applies to the primary heat accumulators. The catalyst chambers are preferably also made refractory and an excellent packing is made by impregnating brick fragments with a suitable catalyst, one of the best being a soluble iron salt which in use becomes converted to the oxide. However, selection of any acceptable catalyst forms no part of my process many excellent proprietary materials being available.

In the event that a brick packing impregnated with a soluble salt of some metal be selected then the mass of said packing also converts the catalyst chamber into a type of supplementary heat accumulator, the gradient of highest efficiency of said catalyst moving forwards and backwards with each reversal of flow.

When the fierce heat of the combustion chamber has been tempered by passage through the primary accumulators and through the catalyst chambers then a metallic packing, such as turnings or other subdivided material with much surface, will be advantageous in the secondary accumulators. Stainless steel, Monel metal, and other resistant metals or alloys are all acceptable. Scrubber and blower, of course, are conventional.

Having thus fully described my process, I claim:

1. The method of making hydrogen which comprises; passing a gaseous fluid consisting essentially of hydrogen, nitrogen and steam, alternately in opposite directions, through a body of carbon maintained at reaction temperatures by the controlled admission of air, at such a rate that the carbon dioxide produced shall at no time exceed three times the weight of the resident hydrogen; storing a portion of the sensible heat in said gaseous fluid as it leaves the combustion chamber in two alternate heat accumulators on each reversal of flow; removing the carbon dioxide thus produced from said fluid after said cooling through the heat accumulators; splitting off a fraction of said gaseous fluid as an accepted fraction; conveying stored heat to the remaining fraction by passage through the alternate heat accumulator and then re-cycling said fraction to the body of carbon originally cited.

2. The method of making hydrogen set forth in claim 1, with the added step that after partial cooling by passage through a heat accumulator said gases be next passed over a catalyst suitable for promoting the reaction between CO and steam, the total heat thus generated as well as the heat not abstracted in the former accumulator being stored in a secondary accumulator prior to removal of CO_2.

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