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# UNDERGROUND HYDROGENATION OF OIL

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This invention is concerned with a novel method of recovering petroleum from underground reservoirs thereof. More particularly, it relates to a means by which petroleum is rendered less viscous and, hence, more readily recoverable by other known methods, such as, of example, gravity drainage or by water flooding.

Specifically, the process of my invention contemplates moderate heating of a tar sand or a reservoir of viscous oil having an API gravity not substantially in excess of about 10° and thereafter contacting the resulting heated oil with hydrogen. The hydrogen remains in contact with the hot oil for an extended period of time whereby under reservoir conditions of pressure and temperature hydrogenolysis of the oil occurs causing a marked reduction in molecular weight and viscosity of the oil.

Some of the largest known liquid petroleum deposits in the world are the Athabasca tar sands located in northern Alberta. It has been estimated that this area alone contains approximately three hundred billion barrels of oil. Other huge deposits of a similar nature are to be found in various parts of the United States and in Venezuela. Owing, however, to the highly viscous nature of these deposits, their production has presented an extremely difficult problem. Numerous proposals have been made in an effort to recover such material including, for example, processes involving mining the tar and thereafter centrifuging it in the presence of certain solvents and surface active agents to separate the tar from the sand with which it is associated. Also, attempts have been made to recover oil from the tar sand by subjecting the latter to treatment with hot water and separating the resulting upper oil layer. These and other methods which have been used, however, all require large labor and capital expenditures rendering such procedures economically unattractive.

Underground combustion as a means of recovering deposits of this type has likewise been suggested. In general, however, the very high differential pressures that must be applied between input and producing wells to recover the oil presents an extremely difficult problem. Frequently, the pressures that must be applied to shallow reservoirs of low permeability, i.e., less than 100 millidarcies, are higher than can be applied economically and/or without causing uncontrolled fracturing of the formation which would lead to channeling and bypassing.

Conventional underground combustion, i.e., an operation in which the combustion zone is propagated from a point near the face of an injection well toward a producing well, is impossible with heavy viscous hydrocarbons of the type contemplated herein. This is for the reason that the hot portion of the reservoir rock yielding the heavy oil lies between the injection well and the burning zone. In this zone the viscosity of the oil is at a minimum; however, as the pressure of the system forces the oil toward the producing well, the oil decreases in temperature to that of the unburned portion of the reservoir. Eventually, resistance to flow through the reservoir to the producing well becomes so great that combustion can no longer continue because it is impossible to supply air at a satisfactory rate to the burning zone.

While reverse combustion, as described and claimed in U.S. Patent 2,793,693 Morse, has been employed on a limited scale to recover oil under conditions of low reservoir permeability, the successful application of this process appears to be related to a restricted class of reservoirs. The process of my invention serves to widen the scope of applicable situations to which the basic process of the Morse patent, referred to above, may be used.



While a certain amount of oil can ordinarily be recovered by reverse combustion, such reservoirs usually must be subjected to a forward combustion process—after reverse combustion, to obtain any appreciable recovery, as described and claimed in U.S. Patent No. 3,174,544 by Frank E. Campion et al.

By the present invention, I am able to recover oil from such reservoirs, and after reverse combustion thereof or otherwise heating the viscous oil or tar, by contacting the hot petroleum with hydrogen in the absence of added hydrogenation catalyst. At prevailing pressures, e.g. 600 to 1000 p.s.i., and temperatures, e.g. 350° to 900° F., hydrogenolysis of the oil in place can be effected, causing a decrease in oil viscosity and thus rendering possible the recovery of such oil by conventional secondary methods. To generate temperatures of 350° to 900° F. during reverse combustion, an air flux of about 30 to about 100 s.c.f.h. per square foot should preferably be used. Ordinarily, temperatures of the order of from about 400° to about 550° F. are preferred. In any event, the temperature should be below that at which excessive decomposition of petroleum occurs.

The process of my invention is primarily applicable to reservoirs having a low effective initial or relative permeability. By the expression "low effective initial permeability," I mean a formation in which conventional forward combustion alone cannot be carried out owing to the fact that the oil temporarily reduced in viscosity in the combustion zone increases in viscosity when it contacts cold reservoir rock on its way to the producing well and hence its resistance to flow through the rock becomes so great that it is either uneconomical or impossible to continue air injection. Stated otherwise, forward combustion alone is considered feasible only when the flow capacity of the reservoir in millidarcy feet is greater than about 30 times the oil viscosity in centipoises. The aforesaid expression, as applied to situations where forward combustion alone cannot be used to recover oil in commercial quantities refers to reservoirs where the maximum air injection rate is insufficient to produce a combustion zone temperature of about 800° F.

Actually, the process of my invention may be employed to advantage in reservoirs having an initial effective permeability so low that substantially none of the hydrocarbon in place is distilled or cracked during reverse combustion. For example, in tar sands in which the initial effective permeability is so low that the maximum amount of air injected results in a peak temperature of not more than about 300° to 400° F., no oil is produced during the reverse combustion phase of the process.

It should be pointed out that forward combustion also could be employed, i.e., generally as taught in the aforesaid U.S. Patent No. 3,174,544, subsequent to treatment of hot oil in the reservoir with hydrogen. However, when the oil is subjected to hydrogenolysis between the reverse and forward combustion steps in accordance with my invention, oil of better quality is obtained in increased amounts and at lower air-oil ratios. The quality of the oil thus produced is higher because it is not only subjected to treatment with hydrogen under conditions favoring a break-down of molecules into smaller, more volatile ones, but the oil thus treated is further improved by the cracking conditions to which it is subjected during the forward combustion step. Increased oil recovery and lower air-oil ratios as provided by the process of my invention are interrelated factors. Thus, in forward combustion it is known that the combustion front does not move toward the producing well until all carbonaceous

material at the front has been burned. In the case of heavy viscous oils and tars, the front moves very slowly, owing to the high proportion of components therein having a low volatility. Because of the slowness with which the front moves under such conditions, more air is required to burn this relatively large proportion of a heavy oil and tar. With a high proportion of these heavy components converted into lighter, lower boiling fractions, less air is required because the combustion front moves at a faster rate since the quantity of heavier hydrocarbons serving as fuel for the combustion front has been decreased. This also makes for increased production due to the tendency of increased quantity of lower boiling hydrocarbons to be displaced ahead of the combustion front.

While hydrogenation rates as contemplated by my invention are far below the lowest economic limits insofar as concerns above-ground operations, such rates are of no appreciable importance because of the long residence times available, thereby assuring the occurrence of hydrogenolysis which, in turn, results in a marked reduction of viscosity and in upgrading in quality of the oil.

The hydrogen used in this process may, of course, be obtained from a variety of sources. However, in general, I prefer to prepare it by well known reforming methods. As the fuel for the manufacture of hydrogen by such methods, a liquid fraction from the produced oil, or the gas or coke produced from thermal cracking of the viscous oil or tar may be used. Cracking occurs to some extent in the formation, depending, of course, on the temperature. However, the heavier oil fractions may be separated from the oil produced and used as a reformer fuel in a known manner. Actually, an impure hydrogen stream can be employed such as that obtained by reforming without carbon dioxide removal. In some instances, carbon dioxide removal or partial removal by any of the well known methods may be advisable, however. Steam required in the reforming process may be derived by burning the coke or tar residue fraction resulting from cracking of the oil in place at the higher end of the aforesaid temperature scale, i.e., from about 600° to about 900° F. This reformer product, which contains from about 35 to about 65 percent hydrogen can usually be injected into the formation without further purification since the impurities present do not interfere to any substantial degree with the desired hydrogenolysis reaction. As a specific project progresses, it will be realized that one portion of a field can be produced while another portion is taking hydrogen into the oil-bearing formation while possibly still another section of the field is undergoing forward combustion. The gas from producing wells under these conditions contains an appreciable amount of hydrogen together with light gaseous hydrocarbons. This gaseous product can be used as reformer feed to produce additional hydrogen for the process.

The hydrogen employed may, if desired, be introduced into the oil-bearing formation via the producing well and in that way avoid the possibility of contacting free oxygen. Introduction of hydrogen down the injection well also may be practiced. However, the well and piping system should first be purged with a flue gas, recycled product gas, or the equivalent.

The pressure used in carrying out the process of my invention may vary widely, depending on a number of conditions, such as the permeability of the reservoir, the combustion zone temperature, air injection rates, etc. In general, it may be said that the pressure employed should exceed that of the reservoir, but should not be sufficiently great to cause uncontrolled fracturing of the formation and undesirable channeling and bypassing of the injected air. Higher pressures, however, favor a more complete hydrogenolysis of the heavier hydrocarbon fractions. Pressures of from about 300 to about 1200 p.s.i. are typical of those that may be employed.

As previously indicated, hydrogen or hydrogen-containing gas should ordinarily be allowed to remain in contact with the viscous oil or tar at reservoir conditions resulting from reverse combustion or an equivalent heat treatment until samples taken periodically from the producing well or wells show that the produced oil has a maximum viscosity of about 300 centistokes at reservoir conditions. Depending on the conditions of the reservoir and the characteristics of the oil, the time of contact of hydrogen with the oil may vary widely, for example, from about 1 month to 6 months or a year, or even longer. Generally speaking, the hydrogen-containing gas should be introduced into the formation, typically at the rate of 400,000 cubic feet per day per injection well, until a breakthrough of hydrogen is detected in one or more of the producing wells. Otherwise stated, the amount of hydrogen to be employed, in general, may correspond to about the hydrocarbon pore volume of the reservoir involved at reservoir conditions. This operation in itself, depending on the size of the project, may require a period of several months. When the composition of the produced gas is at least about 25 percent of injected reformer gas or the substantial equivalent thereof, the producing wells should be shut in. Thereafter, hydrogen injection—or gases containing appreciable quantities of hydrogen, should be in an amount sufficient to maintain adequate pressure at the injection well or wells. These conditions should be maintained for at least about one month. While such periods of time for the hydrogen to remain in contact with the viscous oil are generally preferred, an improvement in quality of the oil can be secured if production is begun shortly, i.e., within a matter of hours, after hydrogen is first detected in the producing well. Thereafter, forward combustion can be continued using the original injection wells or the original producing wells for air injection. It will be realized that under such circumstances large quantities of hydrogen will remain in the reservoir in contact with the oil at high temperatures, particularly near the combustion front. This condition, coupled with substantial reservoir pressures, assists materially in effecting hydrogenolysis of the oil in accordance with my invention.

In some cases sufficient reduction in viscosity of the oil on introduction of hydrogen into the reservoir and prolonged contact therewith may occur to such an extent that a forward combustion step is unnecessary. In instances of this sort, the wells can be produced as previously indicated by such methods as gravity drainage, steam drive, or by water flooding. Even in cases of this sort, however, a large proportion of the hydrogen still remains in contact with oil or tar under producing conditions to have a very substantial effect on reduction of oil viscosity.

In regard to the relationship of oil recovery efficiency to combustion zone temperature used in reverse combustion, it has been observed that below about 400° F. very little oil is produced. Production then increases rapidly with increasing temperature to a maximum at about 900° F. In the case of Athabasca Tar Sand very little, if any, of the tar vaporizes at temperatures below 400° F. Oil recovery then increases with temperature until cracking occurs at about 650° to 700° F. At temperatures below cracking temperature, oil recovery by reverse combustion is controlled by the distillation characteristics of the tar and increases with temperature up to a level of about 900° F. At temperatures appreciably above 900° F., more of the oil or tar in place is required to burn to support the combustion zone. Accordingly, above temperatures of 800° to 900° F., the recovery declines because of the additional amount of hydrocarbon necessary for the combustion step.

For a better understanding of my invention the following example is included:

*Example*

Two stainless steel cells 18 inches long and 2 inches I.D. were packed with Athabasca tar sand to a density

of about 118 lbs./ft.$^3$. The sand contained 16 wt. percent tar. In cell A, the total charge of tar sand was 3615 gms. while in cell B the weight of the charge was 3571 gms. Both cells were heated to a temperature of 500° F. and cell A was subjected to a nitrogen pressure of 500 p.s.i.g., with cell B being subjected to a hydrogen pressure of 500 p.s.i.g. Both cells were left under these conditions of temperature and pressure for a period of one month. The nitrogen pressured cell was used as a control to demonstrate the effect of heat alone on the characteristics of the tar. After the one-month period, the charges of tar sand from each of the cells were separated, extracted with benzene, the latter removed from the resulting extract and the viscosity of the residue determined. The results obtained are shown below.

TABLE I

| Tar Sample | Viscosity, Centistokes | Temperature, ° F. |
|---|---|---|
| Untreated tar | 418 | 300 |
| Do | 13 | 400 |
| Nitrogen treated (Cell A) | 535 | 240 |
| Do | 78 | 300 |
| Hydrogen treated (Cell B) | 180 | 115 |
| Do | 34 | 300 |

The above data clearly show the favorable effect of hydrogen under the conditions contemplated by my invention. Obviously, heat alone or an inert gas at elevated temperature, do not produce the desirable result that is obtained when crude oil is left in contact with hydrogen over an extended period.

While the present description has been directed to a method for improving the quality of tars and viscous oils, it is to be emphasized that my invention is likewise suited to upgrading oils of lower initial viscosity. Any crude oil coming in contact with hydrogen under the conditions contemplated herein will be substantially improved in quality. With the application of my invention to crudes of lower viscosity it will also be evident that under proper conditions forward combustion could be used to produce the required reservoir temperature. It is likewise possible that forward combustion could be used as a means of recovering the oil after treatment with hydrogen has been completed.

In the present description and claims the term "viscous oil" as used herein is intended to include both tars and heavy oils as previously defined.

I claim:

1. In a method for recovering petroleum from an underground deposit thereof penetrated by an injection well and a producing well and wherein said deposit has a low effective initial permeability and has been heated to a temperature of from about 350° to about 900° F. by means of reverse combustion, the improvement which comprises:

injecting a gaseous hydrogen-containing mixture via said injection well into said deposit in the absence of added hydrogenation catalyst while at a temperature within the aforesaid range,

continuing said injection step until at least about 25 percent of the gas flowing from said producing well is derived from said injected mixture,

shutting in said producing well and maintaining sufficient pressure at said injection well to retain the hydrogen in said mixture in contact with said petroleum for a period of at least about one month,

thereafter subjecting said deposit to forward combustion and recovering oil from said deposit via one of said wells.

2. The process of claim 1 in which the hydrogen employed is produced by reforming the gaseous fraction recovered from one of said wells.

3. A method for upgrading the quality of a viscous oil in an underground deposit thereof penetrated by an injection well and a producing well and wherein said deposit has a low effective initial permeability and has been heated to a temperature of from about 350° to about 900° F. by means of reverse combustion, the improvement which comprises:

injecting a gaseous hydrogen-containing mixture via said injection well into said deposit in the absence of added hydrogenation catalyst while at a temperature within the aforesaid range,

continuing said injection step until at least about 25 percent of the gas flowing from said producing well is derived from said injected mixture,

shutting in said producing well and maintaining sufficient pressure at said injection well to retain the hydrogen in said mixture in contact with said petroleum for a period of at least about one month,

thereafter subjecting said deposit to forward combustion and recovering oil from said deposit via one of said wells.

4. A method for upgrading the quality of a viscous oil in an underground deposit thereof penetrated by an injection well and a producing well and wherein said deposit has a low effective initial permeability and has been heated to a temperature of from about 350° to about 900° F. by means of reverse combusion, the improvement which comprises:

injecting a gaseous hydrogen-containing mixture via said injection well in said deposit in the absence of added hydrogenation catalyst while at a temperature within the aforesaid range,

continuing said injection step until at least about 25 percent of the gas flowing from said producing well is derived from said injected mixture,

shutting in the producing well and maintaining sufficient pressure at the injection well to retain said mixture in contact with said oil for a period of at least about one month,

thereafter subjecting said deposit to forward combustion, and

recovering oil from said deposit via one of said wells.

5. A method for upgrading the quality of a viscous oil in an underground deposit thereof penetrated by an injection well and a producing well and wherein said deposit has a low effective initial permeability and has been heated to a temperature of from about 350° to about 900° F. by means of reverse combustion, the improvement which comprises:

injecting hydrogen via said injection well into said deposit in the absence of added hydrogenation catalyst while at a temperature within the aforesaid range,

continuing said injection step until at least about 25 percent of the gas flowing from said producing well is derived from said injected mixture,

shutting in the producing well and maintaining sufficient pressure at the injection well to retain said mixture in contact with said oil for a period of at least about one month,

thereafter subjecting said deposit to forward combustion, and

recovering oil from said deposit via one of said wells by means of gravity drainage.

6. A method for upgrading the quality of a viscous oil in an underground deposit thereof penetrated by an injection well and a producing well and wherein said deposit has a low effective initial permeability and has been heated to a temperature of from about 350° to about 900° F. the improvement which comprises:

injecting hydrogen via said injection well into said deposit in the absence of added hydrogenation catalyst while at a temperature within the aforesaid range,

continuing said injection step until hydrogen is detected in the effluent from said producing well,

thereafter discontinuing injection of hydrogen,
shutting in the producing well and maintaining sufficient pressure at the injection well to retain said mixture in contact with said oil for a period of at least about one month,
next subjecting said deposit to forward combustion, and
recovering oil from said deposit via one of said wells.

**7.** The process of claim **6** in which the hydrogen employed is produced by reforming a fluid fraction recovered from said producing well.

**8.** In a method for recovering petroleum from an underground deposit thereof, said deposit being penetrated by a producing well and an injection well, and wherein said deposit has a low effective initial permeability and has been heated by means of reverse combustion to a temperature of from about 350° F. to a level below that at which excessive decomposition of said petroleum occurs, the improvement which comprises:
injecting hydrogen into said deposit in the absence of added hydrogenation catalyst via said injection well while at a temperature within the aforesaid range,
continuing injection of hydrogen into said deposit until hydrogen can be detected in said producing well,
shutting in said producing well and maintaining sufficient pressure at said injection well to retain said hydrogen in contact with said petroleum for a period of at least about one month, and
thereafter recovering oil from said deposit via one of said wells by means of forward combustion.

**9.** A method for upgrading the quality of viscous oils having a viscosity of at least about 10,000 centistokes in an underground deposit thereof penetrated by an injection well and a producing well and wherein said deposit has a low effective initial permeability and has been heated to a temperature of from about 350° to about 900° F by means of reverse combustion, the improvement which comprises:
injecting hydrogen-containing gas via said injection well into said deposit in the absence of added hydrogenation catalyst while at a temperature within the aforesaid range,
continuing said injection step until hydrogen can be detected in the gases flowing from said producing well,
discontinuing injection of said gas,
shutting in the producing well and maintaining sufficient pressure at said injection well to retain said hydrogen-containing gas in contact with said petroleum until a sample of produced oil has a maximum viscosity of about 300 centistokes at reservoir conditions, and
thereafter producing oil of reduced viscosity from one of said wells by means of forward combustion.

**10.** A method for upgrading the quality of a viscous oil having an API gravity of not more than about 10° in an underground deposit thereof penetrated by an injection well and a producing well and wherein said deposit has a low effective initial permeability and has been heated to a temperature of from about 350° to about 900° F. by means of reverse combustion, the improvement which comprises:
injecting hydrogen-containing gas via said injection well into said deposit in the absence of added hydrogenation catalyst while at a temperature within the aforesaid range,
continuing said injection step until hydrogen can be detected in the gases flowing from said producing well,
discontinuing injection of said gas,
shutting in the producing well and maintaining sufficient pressure at said injection well to retain said hydrogen-containing gas in contact with said petroleum until a sample of produced oil has a minimum viscosity of about 300 centistokes at reservoir conditions, and
thereafter producing oil of reduced viscosity from one of said wells by means of forward combustion.



**11.** A method for upgrading the quality of viscous oils having a viscosity of at least about 10,000 centistokes in an underground deposit thereof penetrated by an injection well and a producing well and wherein said deposit has a low effective initial permeability and has been heated to a temperature of from about 350° to about 900° F., by means of reverse combustion the improvement which comprises:
injecting hydrogen-containing gas via said injection well into said deposit in the absence of added hydrogenation catalyst while at a temperature within the aforesaid range,
continuing said injection step until hydrogen can be detected in the gases flowing from said producing well,
discontinuing injection of said gas,
shutting in the producing well and maintaining sufficient pressure at said injection well to retain said hydrogen-containing gas in contact with said petroleum until a sample of produced oil has a maximum viscosity of about 300 centistokes at reservoir conditions, and
thereafter producing oil of reduced viscosity from one of said wells by introducing into said deposit via the other of said wells a fluid selected from the group consisting of steam and water.

**12.** A secondary recovery method for recovering residual liquid hydrocarbons from a permeable underground formation traversed by injection and production wells which comprises:
(a) continuously subjecting the hydrocarbons in the formation between said wells to controlled in-situ combustion in which a combustion front having a temperature of between about 400° F. and about 850° F. is propagated through said formation to convert a portion of the hydrocarbons therein to hydrogenatable, residual carbonaceous material;
(b) contacting the residual carbonaceous material with a hydrogenating gas at a pressure exceeding about 500 p.s.i.g. and a temperature of from about 400° F. to about 950° F. for a period sufficient to substantially lower the viscosity of said residual carbonaceous material by injecting hydrogenating gas through a first one of said wells;
(c) recovering said hydrogenated residual material from the formation through a second one of said wells.

**13.** A method for hydrogenating subterranean hydrocarbons in place in a formation traversed by injection and production wells which comprises:
(a) continuously subjecting the hydrocarbons in said formation between said wells to controlled reverse in-situ combustion in which a driving, combustion-supporting gas is passed through the formation toward a producing well, said combustion front is propagated in a direction opposite to the direction of movement of said combustion-supporting gas, and the rate of introduction into the formation of said combustion-supporting gas is controlled to maintain the temperature at the combustion front between about 400° F. and about 850° F.;
(b) injecting a hydrogenating gas into said formation through one of said wells to hydrogenate the residual carbonaceous material remaining in the formation following said controlled reverse in-situ combustion; and then
(c) recovering said hydrogenated residual material from the formation through one of said wells.

**14.** The method claimed in claim **13** wherein said hydrogenating gas is hydrogen and the hydrogen is injected

into the formation until a pressure exceeding 1000 p.s.i.g. is reached in the formation.

15. A process for recovering residual carbonaceous materials remaining in a formation traversed by injection and production wells after passing a continuous reverse in situ combustion front controlled to have a temperature in the range of about 400° F. to about 850° F. through the formation between said wells, which process comprises:

(a) injecting a hydrogenating gas into the formation through one of said wells at a pressure exceeding 1000 p.s.i.g. while maintaining the temperature in the formation between 400° F. and 850° F. so as to hydrogenate said residual carbonaceous materials; and

(b) recovering hydrogenated residual material from the formation through one of said wells.

16. A process as claimed in claim 15 wherein the hydrogenated residual material is removed by water flooding the formation.

17. A process as claimed in claim 15 wherein the hydrogenated residual material is recovered by passing a combustion front through the formation.

18. The method of recovering viscous crude oil from a subterranean formation which comprises:

(a) penetrating the formation with a pair of wells spaced from each other and adapted for use as an injection well and a producing well;

(b) initiating combustion of the oil in said formation adjacent the producing well;

(c) continuously injecting a combustion-supporting gas into said formation from said injection well to maintain the combustion of oil in said formation and propagate the combustion front at a temperature of



between about 400° F. and about 850° F. in a direction away from said producing well and toward said injection well; then, following the movement of the combustion front from adjacent the producing well to adjacent the injection well;

(d) injecting a hydrogenating gas into the formation via one of said wells while at least partially shutting in the other of said wells to build up the gas pressure in said formation to greater than 500 p.s.i.g.

(e) maintaining the hydrogenating gas in the formation at a pressure exceeding 500 p.s.i.g. until maximum hydrogenation of the residual carbonaceous material in the formation is achieved; then

(f) recovering the hydrogenated carbonaceous material from the formation through one of said wells.

19. The method claimed in claim 18 wherein said hydrogenating gas is hydrogen.

20. The method claimed in claim 18 wherein the hydrogenating gas pressure in said formation is built up to in excess of 1000 p.s.i.g. and said pressure is maintained until maximum hydrogenation of the residual carbonaceous material in the formation is achieved.

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