High-Temperature Reactors for Underground Liquid-Fuels Production with Direct Carbon Sequestration

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Abstract – *The world faces two major challenges: (1) reducing dependence on oil from unstable parts of the world and (2) minimizing greenhouse gas emissions. Oil provides 39% of the energy needs of the United States, and oil refineries consume over 7% of the total energy. The world is running out of light crude oil and is increasingly using heavier fossil feedstocks such as heavy oils, tar sands, oil shale, and coal for the production of liquid fuels (gasoline, diesel, and jet fuel). With heavier feedstocks, more energy is needed to convert the feedstocks into liquid fuels. In the extreme case of coal liquefaction, the energy consumed in the liquefaction process is almost twice the energy value of the liquid fuel. This trend implies large increases in carbon dioxide releases per liter of liquid transport fuel that is produced.*

It is proposed that high-temperature nuclear heat be used to refine hydrocarbon feedstocks (heavy oil, tar sands, oil shale, and coal) "insitu", i.e., underground. Using these resources for liquid fuel production would potentially enable the United States to become an exporter of oil while sequestering carbon from the refining process underground as carbon. This option has become potentially viable because of three technical developments: precision drilling, underground isolation of geological formations with freeze walls, and the understanding that the slow heating of heavy hydrocarbons (versus fast heating) increases the yield of light oils while producing a high-carbon solid residue. Required peak reactor temperatures are near 700°C—temperatures within the current capabilities of high-temperature reactors.

I. INTRODUCTION

The world faces two major challenges: (1) reducing dependence on oil from unstable parts of the world and (2) minimization of greenhouse gas emissions. A strategy to produce sufficient oil to ensure oil independence while reducing the carbon emissions to the atmosphere is proposed that uses high-temperature reactors. The peak high-temperature reactor temperatures that would be required are near 700°C—a temperature that has been exceeded by all demonstration high-temperature reactors and that does not require development of new reactor technologies. The process uses heat from the reactor and thus does not require development of heat-to-electricity or heat-to-hydrogen systems.

The traditional methods used to produce transport fuels in refineries and the alternative strategy of underground refining are described. The concept of underground refining has existed for decades. What has changed is that the advancing technology may now make it a practical economic process; however, underground refining requires massive quantities of heat. If conventional fossil energy sources are used to provide that heat, there will be large increases in greenhouse gas releases to the atmosphere unless underground sequestration of carbon dioxide is undertaken. However, carbon dioxide sequestration is not a fully demonstrated technology. Equally important, the sequestration technology requires the appropriate geology that may or may not be in the same parts of the country that have appropriate geologies for underground refining. If nuclear energy provides this heat, carbon dioxide emissions from the production of transport fuels can be drastically reduced.

II. TRADITIONAL CRUDE-OIL REFINING

The fuel cycle for traditional liquid fuels includes obtaining the feedstock; converting that feedstock to liquid fuels; transporting the liquid fuels to the user; and burning the liquid fuel in a car, truck, or airplane. Each step consumes energy. Figure 1 shows the greenhouse gas releases per vehicle mile[1] from a diesel-powered SUV for each step in today's fossil fuel cycle for liquid fuels. The greenhouse gas releases are roughly proportional to the energy consumed in each step. With the production of liquid fuels such as diesel from fossil fuels, the total fuel-cycle energy consumption and carbon dioxide releases to the atmosphere are 130 to 200% of the energy consumption and carbon dioxide released from the vehicle.

The process of extracting high-quality sweet (low-sulfur) crude oil (such as Wyoming sweet crude oil), converting it to diesel fuel, and transporting the diesel fuel to the fuel pump consumes relatively little energy and releases relatively small quantities of carbon dioxide. In contrast, if liquid fuels are made from coal, more energy is consumed in the production process than is available in the final fuel. As the stocks of high-grade crude oil are exhausted and liquid fuel is made from lower-grade resources, much more energy is needed to make a gallon of liquid fuel. The expectation is that there will be major increases in greenhouse gas emissions per unit of gasoline or diesel fuel produced within the next several decades. Greenhouse-gas releases cannot be controlled if this trend continues with existing technologies.

Large refineries are among the largest industrial facilities on earth with capital costs of several tens of billions of dollars each and equally large rates of internal energy consumption. About 7% of the total U.S. energy demand is used within the 149 refineries located in the United States. To understand the potential uses of nuclear energy for liquid fuels production by underground refining, some understanding of the energy demand of a traditional integrated refinery[2–3] is required. Underground refining is based on the same processes used in traditional refining. Figure 2 (left) shows a highly simplified flowsheet of several major components of an integrated refinery.

The one common operation within all refineries is distillation: the primary process used to separate hydrocarbon mixtures into gasoline, diesel, jet fuel, and other mixtures. The first operation in any refinery is the atmospheric distillation column, where the crude oil is heated to ~400°C, most of the oil is vaporized, and various fractions are condensed at different temperatures to produce different intermediate products that are further processed to produce liquid fuels. The heavy oil components that do not vaporize are heated to ~450°C and sent to a vacuum distillation column that operates at low pressure and separates the heavy components into various fractions. These two distillation operations typically use 35 to 40% of the energy consumed in a refinery.

The liquid from the bottom of the vacuum column is residual oil (resid). At operating temperatures it is a liquid, but at room temperature it is a solid. It is the component of the crude oil that does not vaporize; instead, if further heated, it would decompose before boiling. It contains all of the solid impurities dissolved in the crude oil. High-value crude oils produce very little resid, whereas heavy oils produce large quantities of resid.

Transport fuels have hydrogen-to-carbon molar ratios approaching 2, whereas lower-value crude oils have much lower ratios. After distillation, the low hydrogen-to-carbon components of the crude oil are mostly in the resid. To convert resid to liquid transport fuels, there are two options: (1) add hydrogen to increase the hydrogen-to-carbon ratio or (2) remove excess carbon. Both are done in large integrated refineries.

- *Hydrocracking*. Hydrocracking processes use hydrogen to convert heavy hydrocarbon molecules (such as resid) into more valuable light hydrocarbons such as gasoline. This process is accomplished by simultaneously splitting larger hydrocarbon molecules into smaller molecules and adding hydrogen to the molecules. This option requires massive quantities of hydrogen, but it maximizes the liquid fuel yields per barrel of oil.

- *Thermal cracking*. Thermal cracking heats the resid and coverts it to more valuable lighter hydrocarbons and petrocoke. Petrocoke is a solid that can be used as a boiler fuel. Thermal cracking (Fig. 2; left) is the brute-force process in which the resid is very quickly heated to ~500°C, exits the heat exchanger, and decomposes into lighter hydrocarbons and petrocoke. Because there is a short time delay between heating the resid and the formation of solid coke, rapid heating allows the resid to exit the heat exchanger before forming coke, which would build up on heat exchanger surfaces and cause heat exchanger failures. Thermal cracking represents a significant high-temperature energy consumer in refineries that process heavy oils.

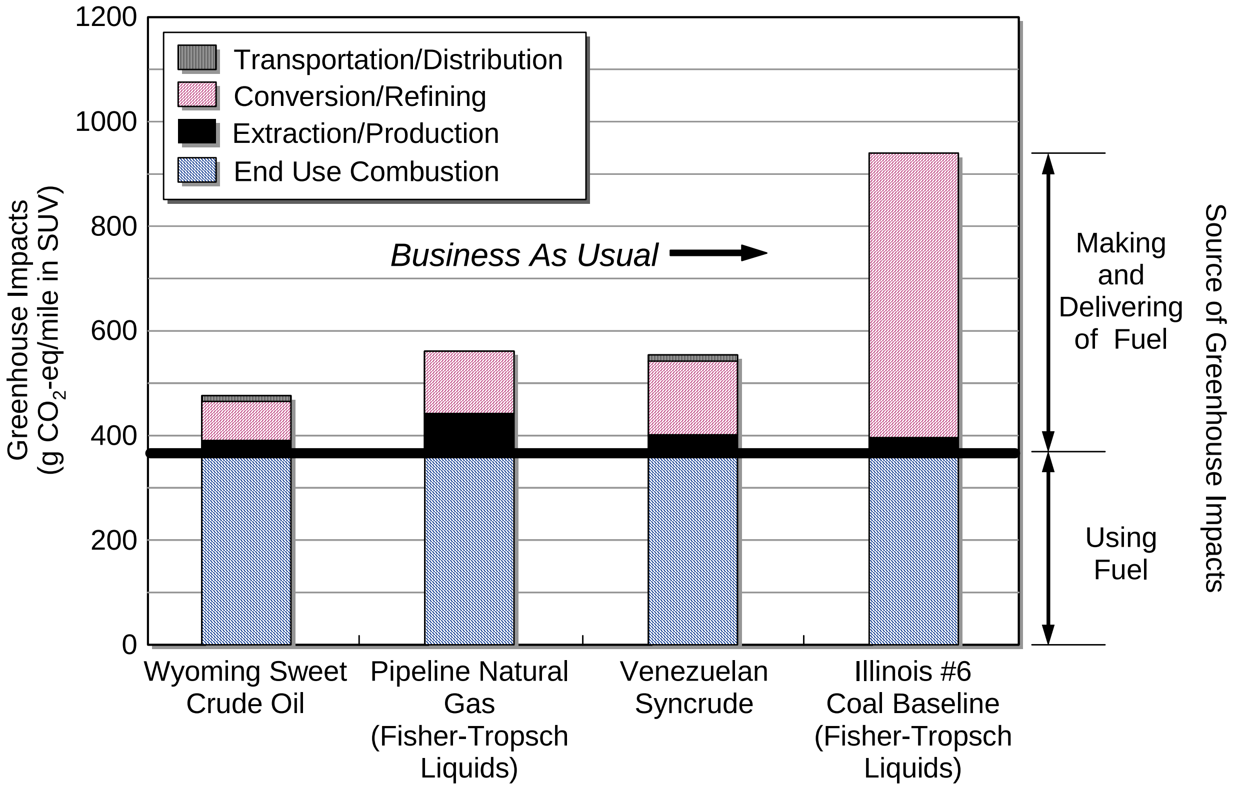

Fig. 1. Greenhouse gas releases per vehicle mile for diesel fuel produced from different sources.[6]

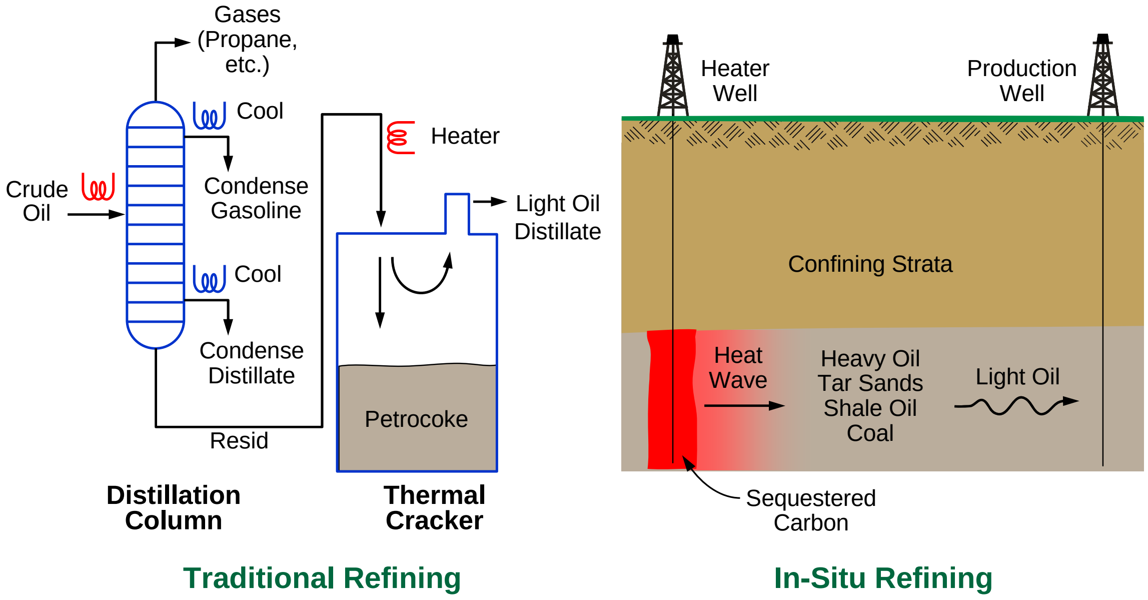

Fig. 2. Distillation and thermal cracking of high-molecular-weight hydrocarbons in a refinery and in an underground hydrocarbon reservoir.

Thermal cracking and hydrocracking have the same goal: production of light hydrocarbons from feedstocks that have a low hydrogen-to-carbon ratio. Thermal cracking obtains the proper hydrogen-to-carbon ratio by removing carbon, whereas hydrocracking obtains the proper hydrogen-to-carbon ratio by adding hydrogen. Both processes result in large quantities of carbon dioxide emissions to the atmosphere. Hydrocracking requires hydrogen that is typically produced by steam reforming of natural gas. In that process, oxygen, natural gas, and water are converted into hydrogen and carbon dioxide. With thermal cracking, the resultant petrocoke is burnt to provide energy, a process that produces carbon dioxide.

III. NUCLEAR REFINERY OPTIONS

As a class, refineries are the largest energy consumers in the United States. The scale of operation of a large integrated refinery exceeds that of a nuclear power plant. Such refineries cost several tens of billions of dollars each and are the largest private industrial facilities on earth. They potentially present two major markets for nuclear energy.

- *High-temperature heat.* For high-quality crude oil, the refinery demand for high-temperature heat is about 10% of the energy value of the final liquid product. Assuming that a refinery uses this quantity of heat, about 7.08 GW(t) would be required for every million barrels of oil that is refined per day. With a national consumption of about 20 million barrels of oil per day, that implies 142 GW(t) of heat. For lower-grade crude oils, more heat is required. To provide that heat, the reactor coolant temperatures will need to be 600 to 700°C. Most of that heat is now provided by natural gas—a premium and expensive fuel.

- *Hydrogen.* Hydrogen is the second prospective market. This market is potentially several times larger. Some understanding of scale can be provided by example. Recently Kuwait announced the construction of another hydrogen plant to provide more hydrogen to their refinery. When fully operational, the rate of energy release if that hydrogen is burned will be ~3000 MW(t).

Because refineries use large quantities of high-temperature heat that is primarily produced by high-cost natural gas, there is commercial interest in using high-temperature reactors to provide that heat. The longer-term option is the production of hydrogen. This is potentially a major market, but there are two limitations. First, the need for alternative sources of oil is not addressed, and secondly, the use of nuclear heat implies retrofitting of existing refineries—a serious constraint at some refinery sites.

IV. NUCLEAR UNDERGROUND REFINING OF FOSSIL FUELS

IV. A. Process Description

There are massive underground resources of fossil fuels that could be converted into liquid fuels but that have been economically unrecoverable with existing technologies. Examples include the following:

- *Old oil fields.* Over half the oil remains in a depleted oil field trapped by capillary forces between grains of sand or within cracks in the rock.

- *Tar sands.* Oil recovery from tar sands has been limited to surface deposits and underground deposits where steam heating can reduce the viscosity of the oil until it flows.

- *Oil shale.* Oil shale is an organic that upon heating is converted to a liquid fuel, some combustible gases, and spent oil shale that contains a solid high-carbon residue.

- *Soft coal.* Soft coal, if heated, is converted to char, a liquid fuel, and some combustible gases.

Starting in the 1970s, researchers began to examine methods to recover oil underground from these fossil deposits. Because of new technologies, concerns about greenhouse gas emissions, and higher oil prices, these technologies are now progressing from the laboratory to field testing, with initial leasing of properties for commercial production.

Conceptually, the technology is simple (Fig. 2, right side). Heat a fossil deposit to high temperature. As the temperatures increase, any volatile hydrocarbons will vaporize (be distilled), move as gases toward recovery wells, condense in the cooler zones, and be pumped out of the ground as a liquid or vapor. This distillation process leaves most impurities behind. While capillary forces can hold liquids in cracks in the rock, gases easily permeate most reservoirs. As the temperature further increases, heavier hydrocarbons will be thermally cracked to produce lighter volatile hydrocarbons that can be recovered. In effect, heating the underground reservoir duplicates the thermal cracking and distillation processes found in a refinery. These processes offer three potential major advantages:

- *Abundant oil*. The fossil resources are very large. For example, the United States has the largest oil shale deposits in the world[4] within the Green River Formation, which covers parts of Colorado, Utah, and Wyoming. The potentially recoverable shale oil resources are between 500 billion and 1.1 trillion barrels. The midpoint estimate is 800 billion barrels, or about three times that of Saudi Arabia. At current rates of U.S. crude oil consumption (~20 million barrels per day), there is sufficient oil to meet the U.S. demand for ~100 years. The resources from other fossil deposits are likely to be similar in scope.

- *Control carbon dioxide emissions*. Unlike the circumstances in a refinery, the solids from an underground thermal-cracking process [petrocoke, char (from coal), spent oil shale, etc.] remain underground sequestered as carbon. While many questions about large-scale sequestering of carbon as carbon dioxide remain unanswered, we know that carbon sequestered as carbon (coal) works. If the heat is provided by an energy source that does not emit carbon dioxide, the result would be low emissions of carbon dioxide from the entire production process because a high-quality crude oil distillate is produced that requires little added refining to produce transport fuels.

- *Reduce toxic materials in the surface environment*. Heavy crude oils and many other hydrocarbon deposits contain significant quantities of toxic heavy metals and carcinogens. Underground refining leaves toxic heavy metals underground and destroys many of the carcinogens in the thermal cracking process.

It may also be possible to undertake hydrocracking by injecting hydrogen into the geology while it is being heated. However, this option has not been investigated. The potential for underground hydrocracking is likely to be strongly dependent upon the particular fossil fuel. Hydrocracking of fossil fuels (1) converts large hydrocarbon molecules into smaller and more valuable liquid fuels and (2) reduces the sulfur content by reacting with the sulfur to produce hydrogen sulfide—a toxic gas that in a refinery is converted to sulfur or sulfuric acid. While fuel desulfurization is desirable in a traditional refinery, it would be highly desirable with underground refining if the sulfur remained underground in the carbon residue with other toxic materials.

IV.B. Shale Oil Processing

In the context of underground production and refining of oil, the most technical progress has been made in the recovery of shale oil. Shale oil deposits represent the most concentrated sources of fossil fuels in the world, as measured in energy content per unit area. Most of the deposits are more than 500 ft thick, with some deposits more than 2000 ft thick and parts of the basin yielding more than 2.5 million barrels of oil per acre. Usable deposits contain from 25 to more than 50 gal/ton of shale oil. This energy content per unit area is far beyond that of any known oil reserve and significantly larger than rich coal deposits such as those of Campbell County, Wyoming, which yield the energy equivalent of less than a half million barrels of oil per acre.

The traditional processes for recovery of shale oil release large quantities of carbon dioxide to the atmosphere and are expensive. These processes involve heating the oil shale to temperatures in excess of 480°C (~900°F) by injecting air and burning some of the organics to produce heat, which then drives the chemical reactions that release the oil from the rock. The process can be conducted aboveground in retorts or underground.

In underground operations, a volume below the retort zone is mined, the shale to be retorted is rubblized by staged explosives, and air is pumped in to burn some of the carbon to produce the required heat. It is currently estimated that for the initial commercial plants, the production costs will be between $70 and $95 per barrel. With additional industrial experience, ultimate costs of $30 to 40 per barrel could probably be achieved.

Since the 1980s, Shell[4–5] and several other companies have been developing new types of in-situ retorting processes. The Shell "In-Situ Conversion Process (ISCPs)" is the closest to commercial deployment. It has been tested on a small scale and is now being scaled up to a precommercial size. While many uncertainties remain, these processes represent a potential revolution in oil shale processing that may produce a premium shale oil with projected production costs of ~$30 per barrel, a cost that is competitive with that of lower-quality crude oil priced in the mid-$20s per barrel. Current oil prices exceed these production costs by large margins.

In the Shell ISCP process, electrical heaters are emplaced in the oil shale and are used to heat a quantity of oil shale through its entire volume. Each acre requires 15 to 25 heaters. After 2 to 3 years, the volume of shale is heated to between 650 and 700°F (370ºC). This slow heating and the relatively low temperature compared with traditional oil shale retorts causes the rock to release oil as well as a gas similar to natural gas. About two-thirds of the energy content is in the oil, and about one-third is in the gas.

The low thermal conductivity of oil shale (poor heat conduction), the economic requirement to heat the bulk rock in a time period of economic interest (several years), and practical necessity of line (well) heating (versus bulk heating) imply peak heater-well temperatures between 600 and 800ºC, with higher temperatures reducing the number of required heaters or decreasing the heating time.

The other technological component is the use of freeze-wall technology to isolate the underground retort from the geological formation. This process, an existing industrial technology, involves drilling wells around the perimeter of the extraction zone and using cooling coils to freeze the groundwater to create a sealed ice wall. The freeze wall (1) allows dewatering of the oil shale and avoids in-leakage of groundwater; (2) keeps the light hydrocarbon products from escaping during ground heating, product extraction, and post-extraction ground cooling; and (3) allows post-treatment of the impacted rock zone to minimize the potential for long-term groundwater contamination.

Approximately 250 to 300 kW(e)-h is required for down-hole heating per barrel of oil. If electricity costs $0.05/kW(e)-h, the power costs are between $12 and $15 per barrel. This value represents over half the production costs. If the electricity is produced from the natural gas from the oil shale with a conversion efficiency of 60%, all of the natural gas produced from the oil shale will be consumed. This gas constitutes one-third the energy content of the hydrocarbons produced in the process. More likely, lower-cost coal would be used to produce the electricity. To produce 100,000 barrels per day, ~1200 MW(e) of electrical generating capacity is required. To meet one-fourth of the U.S. oil demand (~5 million barrels per day), 60,000 MW(e) is required.

An alternative to the use of electricity for shale oil production is the use of high-temperature reactors to produce the high-temperature heat required to heat oil shale (Fig. 3). The heat is transferred from the reactors to the oil shale using liquid-metal or liquid-salt heat-transport loops.[6] The process offers several potential advantages.

- *Energy requirements.* The energy requirements for retorting the oil shale are reduced by a factor of 2 or more. Direct use of high-temperature heat avoids the conversion of heat to electricity (with all the associated losses) and subsequent use of the electricity to produce heat. Thus, expensive electricity is replaced with lower-cost thermal energy.

- *Product recovery*. All of the products from retorting the oil shale are recovered as products and none are burnt to produce the heat required to operate the process.

- *Environmental impacts*. Carbon dioxide and other air emissions are avoided from the production of electricity. The carbon dioxide releases per unit of oil (Fig. 2) will be significantly less than those for other methods used to produce a synthetic crude oil because a light crude oil is produced where the distillation and thermal cracking processes leave impurities and carbon residues underground. The light-oil distillate requires relatively little refining to produce the final product.

The potential viability of using nuclear energy for high-temperature heat is aided by the geological setting of these oil shale deposits. Nuclear reactors are capital intensive, with low operating cost. Good economics requires long-term base-load operations. For this application, heat must be transferred from the reactor to the wellhead. In the thickest oil shale deposits of the Piceance Basin, production of 50,000 barrels of oil per day requires recovery of oil from 15 acres per year and a reactor with a thermal output of ~600 MW(t). In 40 years, the longest heat-transport pipeline from reactor to wellhead (assuming full use of the oil shale, including the area under the reactor) would be <1000 m. Heat can be transported distances of kilometers. Therefore, the distances from reactor to wellhead are sufficiently short that heat transport is practical.

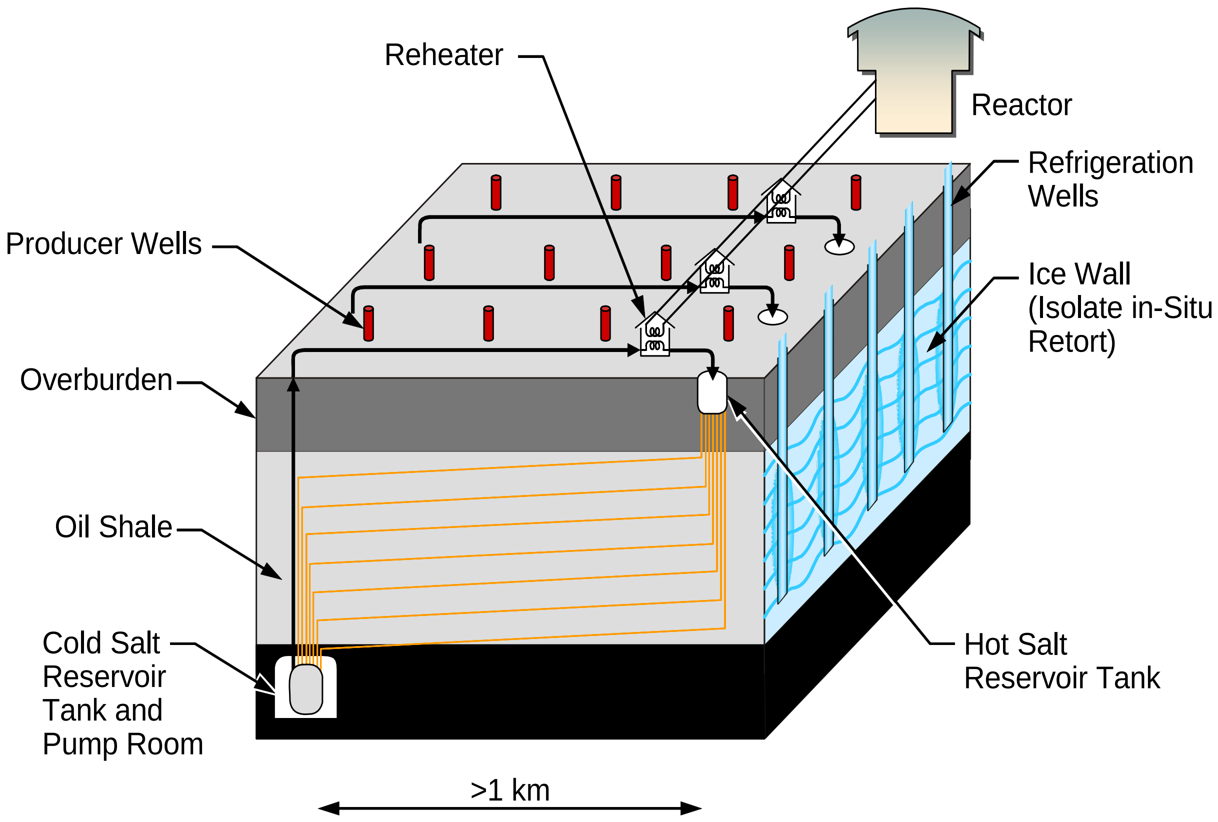

Fig. 3. One configuration for underground heating of oil shale with nuclear heat.

Significant technical challenges are associated with using nuclear energy for oil shale production—including the selection of the appropriate coolant–materials combinations[7] for the heat transfer loops with the development of startup/shutdown procedures. About 12 GW(t) of high-temperature heat would be required to produce a million barrels of oil per day. In practice, the required temperatures would be near 700°C.

IV.C. Other Nuclear Underground Refining Options

The technology for using nuclear heat for recovery of shale oil is potentially applicable to depleted oil fields, tar sands, soft coal, and other hydrocarbon deposits. The economics strongly depends upon the characteristics of the specific fossil fuel and the local geology, which determine the investment in drilling and other activities required per barrel of oil ultimately produced.

In the specific case of coal, multiple processes[8] have been fully or partly developed to heat coal and produce a solid high-carbon char, hydrocarbon liquids, and gases. The classical example is the production of coke that is used, in turn, for the production of pig iron from iron ore. More recently have been work on development of new processes that slowly heat coal to increase yields of liquid hydrocarbons and gases. Various experiments[9] have shown that slow heating (versus rapid heating) of the coal produces larger liquid yields and a higher quality synthetic crude oil. These new processes can be implemented in aboveground and underground locations. The intrinsic characteristic of underground refining using nuclear heat is that it is a slow process because of the low thermal conductivity of rock. Increasing the yield of liquids per ton of coal has major impacts on the economics.

Laboratory and field studies are under way on the use of electric heating of oil sands to recover this oil from deep deposits.[10] While such studies assume electric heating of the rock, the results of many of the studies are applicable to the use of nuclear heat for recovery of these oils. The heating of oil sands and depleted oil fields is somewhat different from the heating of either oil shale or coal. Both oil shale and coal must be heated to thermally crack these molecules into smaller molecules that either flow as liquids or are vaporized and condensed near the production well. In contrast, heating of tar sands or depleted oil fields lowers the viscosity and surface tension of the oil and allows some fraction of the oil to flow as a liquid to the production wells. Any residual oil upon further heating is either vaporized and condenses near the cooler production wells or thermally cracked into more volatile hydrocarbons. Heating deposits to recover oil from tar sands or depleted oil fields will likely require less thermal energy.

IV. CONCLUSIONS

The two major energy challenges for the United States and most of the industrialized world are to (1) replace imported traditional crude oil in our transportation system and (2) reduce greenhouse gas emissions. Recent technology developments indicate the potential of combining underground refining and heat from high-temperature reactors to meet these challenges. Underground refining can potentially recover light-distillate crude oils from oil, heavy oil, tar sands, oil shale, coal, and other hydrocarbon deposits.

While underground refining with nuclear heat will not eliminate carbon dioxide emissions resulting from the use of liquid fuels, it has the potential to drastically reduce carbon dioxide emissions from the production processes that convert fossil fuels into liquid transport fuels. The underground production processes are fundamentally the same processes used today. However, there is a difference. When these processes are conducted underground in the hydrocarbon reservoirs, the residual carbon byproduct is sequestered underground as carbon. The technological challenges are (1) the full commercialization of underground refinery technologies and (2) development of the intermediate heat-transport systems to move heat underground from high-temperature reactors.

REFERENCES

1. J. J. MARANO and J. P. CIFERNO, *Life-Cycle Greenhouse-Gas Emissions Inventory for Fischer-Tropsch Fuels,* Energy, and Environmental Solutions, LLC, for the U.S. Department of Energy National Energy Technology Laboratory (2001).

2. J. H. GARY, G. E. HANDWERK, and M. J. KAISER, *Petroleum Refining: Technology and Economics,* Fifth Edition, CRC Press, Boca Raton, Florida (2007).

3. F. E. SELF, E. L. EKHOLM, and K. E. BOWERS, *Refining Overview—Petroleum, Processes, and Products*, CD-ROM, American Institute of Chemical Engineers (2007).

4. J. T. BARTIS, T. LaTOURRETTE, L. DIXON, D. J. PETERSON, and G. CECCHINE, *Oil Shale Development in the United States: Prospects and Policy Issues*, RAND Corporation (2005).

5. J. BIRGER, "Oil from Stone," *Fortune*, **156** (*10*), 105–116, Nov. 12 (2007).

6. C. W. FORSBERG, "High-Temperature Nuclear Reactors for In-Situ Recovery of Oil from Oil Shale," CD-ROM, Paper 6104, 2006 International Congress on the Advances in Nuclear Power Plants (ICAPP'06), June 4–8, 2006, Reno, Nevada, American Nuclear Society, La Grange Park, Illinois (2006).

7. D. F. WILLIAMS, *Assessment of Candidate Molten Salt Coolants for the NGNP/NHI Heat-Transfer Loop,* ORNL/TM-2006/69, Oak Ridge National Laboratory, Oak Ridge, Tennessee (2006).

8. S. LEE, J. G. SPEIGHT, and S. K. LOYALKA, *Handbook of Alternative Fuel Technologies*, CRC Press (2007).

9. H. R. ZHANG, E. G. EDDINGS, and A. F. SAROFIM, "A Review of Coal Devolatilization with Very Low Heating Rates," CD-ROM, *Proc. American Institute of Chemical Engineers Annual Meeting, Salt Lake City, Utah,* November 4–9 (2007).

10. J. WANG and A. KANTZAS, "Evaluation of Bitumen Production during the Electrical Resistive Heating Process," CD-ROM, *Proc. American Institute of Chemical Engineers Annual Meeting, Salt Lake City, Utah,* November 4–9 (2007).