

Sept. 27, 1960

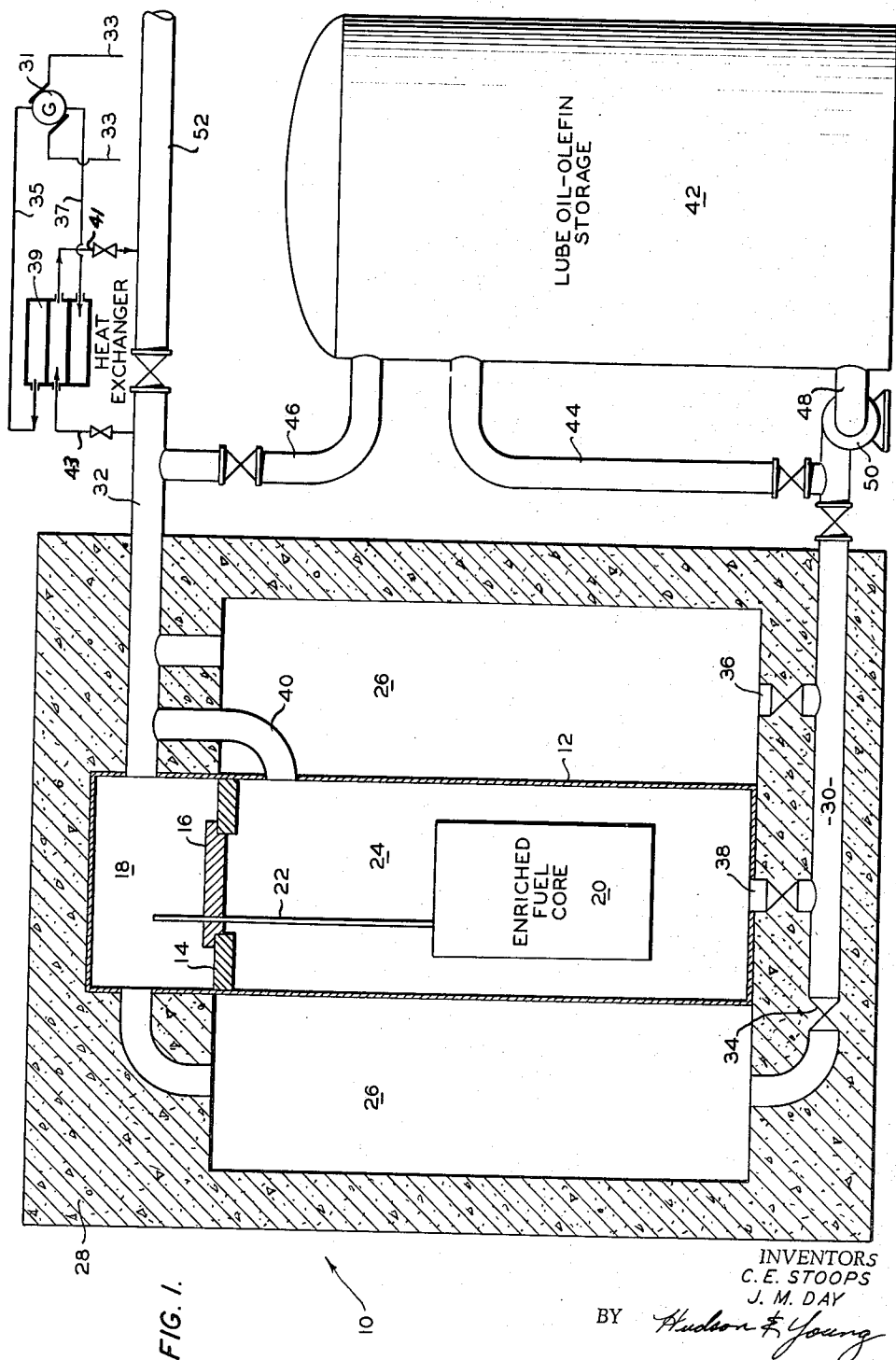
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2,954,334

HIGH VI LUBRICATING OIL AND PROCESS FOR PREPARING SAME

Filed Feb. 25, 1957

3 Sheets-Sheet 1



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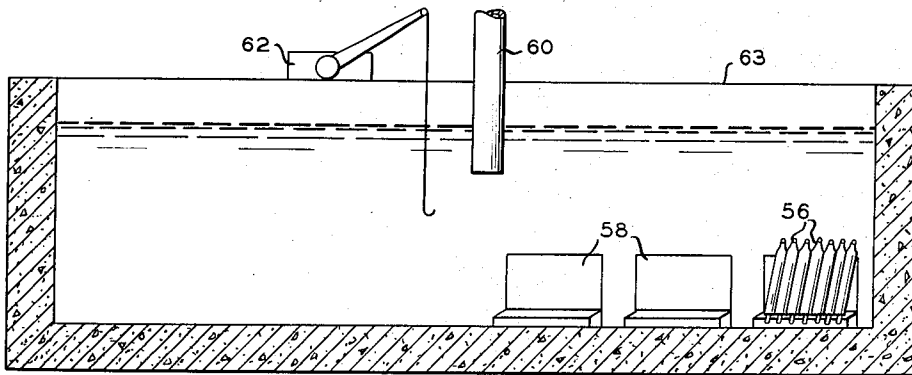


FIG. 2.

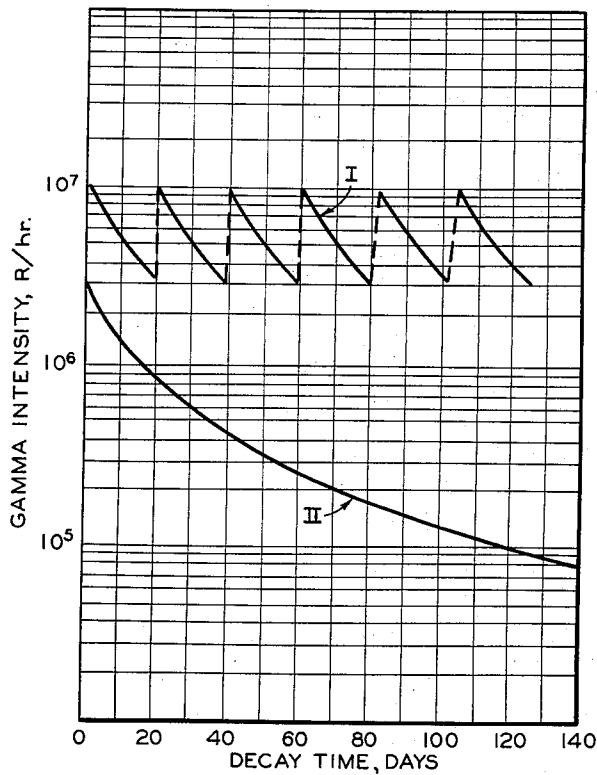


FIG. 6.

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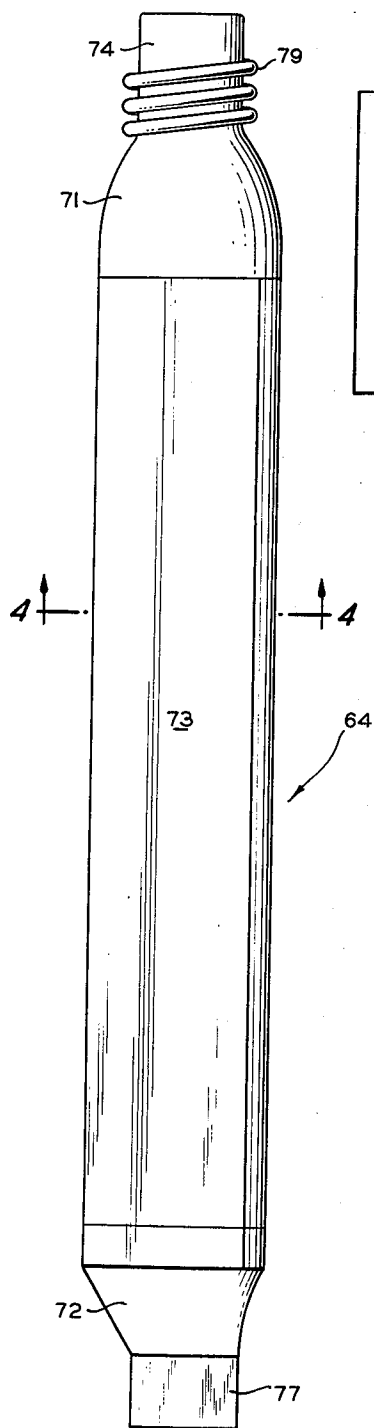


FIG. 3.

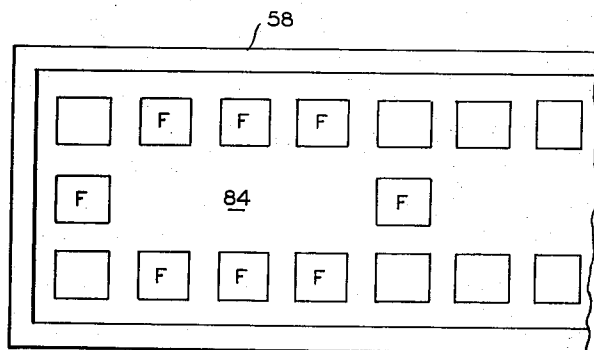


FIG. 5.

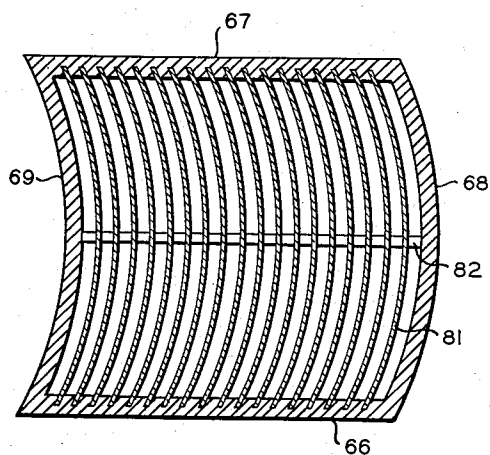


FIG. 4.

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HIGH VI LUBRICATING OIL AND PROCESS FOR PREPARING SAME

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Filed Feb. 25, 1957, Ser. No. 642,103

17 Claims. (Cl. 204—154)

This invention relates to processes for treating lubricating oil with an olefin in the presence of ionizing radiation, and to products of such treatment. In a more specific aspect, the invention relates to employing a mixture of a petroleum lubricating oil and an olefin as a coolant and/or moderator for an atomic reactor, while the mixture at the same time is treated by exposure to radiation generated in the atomic reactor.

It is an object of the invention to provide lubricating oils of improved properties, particularly improved viscosity index. It is another object to provide methods for producing such improved lubricating oils. Other objects of the invention will be apparent from a consideration of the accompanying disclosure.

The utility of oils of high viscosity index for lubrication of modern cars is well known. The need for such high quality oils has resulted in numerous expensive methods for their production. Super refining techniques are helpful, but are limited in the improvement of viscosity index which can be provided. Accordingly, materials which are employed as additives have been developed, known as V.I. improvers. These materials are usually quite expensive to produce.

It has now been found that when a mixture of lubricating oil of petroleum origin, with an olefin is exposed to ionizing radiation until a total dosage of 5×10^6 to 5×10^9 REP is received, a chemical change in the structure of the lube oil is effected which results in a lubricating oil fraction of greatly improved viscosity index. The effect is apparently a result of "grafting" an olefin or an olefin chain onto the molecular structure of the components of the lubricating oil. The total dosage preferred is 10^7 to 10^9 REP.

The development of atomic energy has resulted in the development of terminology peculiar to this particular field. In order to facilitate the understanding of the invention, it is desirable to discuss and clarify some of the terms which are particularly applicable to the atomic energy field. The term "reactor" is applied to the physical means whereby a critical amount of radioactive material is positioned, inclosed, supported and shielded so as to be self-sustaining, i.e., the radioactive material and the environs return as many thermal neutrons per unit of time to the reactor core as are needed to produce the number of fissions per unit time from which they originally come. Obviously, excess reactivity must exist in order to produce useful energy for a reasonably long period of time. This excess reactivity is controlled and introduced as fuel is burned by means of control rods.

The term "fuel" in an atomic reactor includes any of the heavy elements which are capable of undergoing the fission reaction.

Moderators are used so that the neutrons given up by the fission reaction in the fuel core as high energy or fast neutrons, may be slowed down and thereby reduced in energy to the so-called thermal or slow neutron range of energies. This is brought about by elastic collision with the atoms of the moderator, and each elastic collision re-

2

moves a portion of the energy from the neutron in the form of kinetic energy imparted to the molecules of the moderator and which will eventually show up as heat. The best moderators are those having a low capture cross section, low atomic weight and relatively high scatter cross section, that is they are capable of undergoing many elastic collisions and of serving as a medium for neutron diffusion without capturing a neutron in the nucleus. Moderators may be liquids, solids, or gases. However, the latter form lacks sufficient density to be highly effective except at extreme pressures or with infinite space.

A reflector is a blanket of moderating material, surrounding the fuel core (consisting of fuel, moderator, coolant, and such structural material as necessary) which, by moderating and returning neutrons to the fuel core, reduces the amount of fuel necessary for the reactor to support a self-sustaining fission reaction. It also serves as a reservoir of neutrons that can be used for useful purposes, such as the production of radioactive isotopes. In general, moderators and reflectors have the same requirements.

The properties which are necessary and desirable in a moderator are such that hydrogen, carbon, or compounds thereof qualify exceedingly well. None of these will be appreciably activated by irradiation and all have low atomic mass and tend to be very efficient in the absorption of energy from an energetic neutron. They also have a high scatter cross-section so that collisions are frequent. This suggests that organic materials, particularly hydrocarbons, would be good moderating materials. A liquid hydrocarbon material can be used as a coolant as well as a moderator and serve a double purpose.

Although it is impossible in a reasonable length of time to discuss all the ramifications in the choice of moderators and moderator-coolants, the advantages of the use of organic materials are sufficiently obvious to those familiar with the art that the Atomic Energy Commission is expending a large sum of money on an experimental organic moderated reactor. Such an expensive experiment is necessary because of problems stemming largely from the thermal and radiation instability of organic materials in general. A good discussion of some of the advantages of organic moderators and the instability problem and a summary of the Atomic Energy Commission supported laboratory investigations is given by R. G. Bolt and J. G. Carroll (Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, vol. 7, 546-555 (1956)). It is necessary to know the stabilities of many organic materials and to develop techniques for purifying those which are sufficiently stable to use. The latter is necessary because no organic material is completely stable to radiation; the most promising so far have been large aromatic molecules, such as polyphenyls and fused ring aromatics.

The lubricating oils which can be treated according to the invention are those of petroleum origin. Lubricating oils from petroleum are predominantly of high (350-800 or higher) molecular weight. The various types may be named in accordance with the type of crude from which they originate. West Coast, Mid-Continent, and Pennsylvania designate the three major types found in the United States. Pennsylvania crude is predominantly paraffinic in character, Mid-Continent is predominantly asphaltic in character and the West Coast is even more asphaltic with the added character of having considerable amounts of non-hydrocarbon impurities containing sulfur and nitrogen. After careful examination of a Mid-Continent crude, the U.S. Bureau of Standards drew the conclusion that the lube oil fraction is made up of 4,000 *n* constituents. This is based on laborious

distillations made at the Bureau of Standard's Laboratory wherein the lube oil was cut up into 4,000 fractions having different properties and each fraction comprising more than one constituent. It is believed that the factor n is probably a small number greater than one, although its magnitude has not been established. From their findings, it is believed that attempts to speak of lubricating oils in other than very broad terminology is futile.

For good lubricating properties in modern motor cars, an oil must have proper viscosity and high viscosity index. It has been found that these properties are present in fractions composed predominantly of compounds containing a naphthenic nucleus with long paraffinic side chains. The more asphaltic crudes trend toward mixed naphthenic-aromatic nuclei. Long paraffinic side chains mask the effects of the aromatics when measuring such properties as viscosity index and pour point; however, natural formation of the constituents in crude oils appears to favor ring formation at the expense of side chains; and accordingly, the asphaltic crudes contain multi-ring nuclei having varying degrees of unsaturation.

Solvent extraction has largely replaced fractionation for the final separation and preparation of lubricating oil base stocks from crude. The solvents utilized are usually selective for the more aromatic or unsaturated hydrocarbons and therefore yield a raffinate phase richer in the paraffinic and naphthenic stocks, which have desirable lubricating oil properties. As pointed out above, long paraffinic side chains are capable of masking unsaturation; and accordingly, small amounts of unsaturates (aromatic and/or olefinic) are present in lubricating oil base stocks.

The olefins preferred in the process are monoolefins having from 2 to 5 carbon atoms and include ethylene, propylene, 1-butene, 2-butene, isobutylene, 2-methyl-1-butene, 2-methyl-2-butene, 3-methyl-1-butene, 1-pentene and 2-pentene.

In the process of the invention, the lube oil-olefin mixture charged is generally a mixture containing 1 to 2000, preferably 2 to 50 parts by weight of olefins per 100 parts of oil, although higher or lower amounts can be employed.

In the operation of the process of the invention, the source of ionizing radiation with which the oil-olefin mixture is treated can be, for instance, an atomic reactor itself, spent fuel from such a reactor or from any other source of gamma radiation, or other ionizing radiation, of high intensity.

As before stated, an advantageous method of practicing the invention is to effect the irradiation of the mixture while it is serving as a coolant and/or moderator for an atomic reactor. Thus, an advantage of this embodiment of the invention is that there is substituted the reaction mixture of lube oil and olefin for the relatively expensive organic moderators being investigated by the Atomic Energy Commission. A discussion and description of atomic reactors will, therefore, be helpful.

Atomic reactors may be divided into many categories for purposes of discussion. One means of classification is based on the configuration of the fuel elements in the reactor core. Such terms as homogeneous, heterogeneous, and fluidized, are typical examples. By "homogeneous reactor" one denotes a reactor in which the radioactive fuel is in solution in a moderating liquid. In heterogeneous reactors, the fuel is more or less in lumps, clusters of rods and such configurations separated by solid or liquid modifiers. Fluidized reactors, as the name suggests, indicates a reactor in which small particles of moderated fuel are fluidized by non-neutron absorbing gas such as helium and the like. This type of reactor is in the experimental stage and little more information is available. Another classification deals with the type of moderator used, thus we have terms such as light water, heavy water, graphite, and organic, to describe moderated reactors in each of the above fuel configuration categories. Another type of classification relates to the particular type of fuel element used. Examples are natural

uranium, enriched uranium, thorium, and plutonium. Occasionally, such terms as "swimming pool" and "water boiler" are used as further means of identification. These terms refer to similarity of the reactor to well-known structures and phenomena. Another means of identification is based on the use of the reactors, such as power reactors, test reactors, and breeder reactors. Breeder reactors are those in which as much or more nuclear fuel is produced as consumed.

Within the reactor core, the fissioning fuel constantly gives off fast neutrons, gamma rays, and radioactive fission products. The fast neutrons, the prompt gamma rays from the fission process, and the gamma and beta rays from the radioactive fission products are collectively known as ionizing radiation because directly or indirectly in their passage through matter their energy is dissipated by the formation of ion pairs and activated molecules. These ion pairs and activated molecules are responsible for the chemical effects that lead to the degradation of organic compounds. The term ionizing radiation also includes high energy ionic particles such as protons, deuterons, alpha particles, etc., which are not of primary importance in a discussion of reactors, but which might be of importance in the treatment of organic compounds such as lubricating oils. The high energy fission fragments, themselves, are ionizing radiation but are usually retained in the fuel alloy and hence have no effect on the moderator.

The present invention utilizes a lubricating oil-olefin mixture undergoing reaction as the moderator-reflector, as a portion of the moderator, or as a portion of the reflector in any of the experimentally proven reactors, or as a coolant therefore, or both. This is possible because of the low capture cross-section of both the carbon and hydrogen atoms present in the hydrocarbon oil. Graphite, which is pure carbon, is an exceptionally good moderating and reflecting material and hydrogen under high pressure or at low temperatures would also be an excellent moderator or reflector. However, both conditions necessary for its use are extremely difficult to obtain in the reactors under discussion. However, when combined with carbon in the form of hydrocarbons, as in this invention, the resulting material possesses the desirable properties of both carbon and hydrogen and has the advantage that it is a liquid and lends itself to the shape of the container at normal temperatures or pressure, whereas, carbon must be shaped by expensive machine methods and the hydrogen properties as noted above are completely outside the realm of consideration.

In order to provide a more complete understanding of the invention, reference is made to the diagrammatic drawing of which

Figure 1 shows a vertical cross-section of a reactor in which the invention can be practiced.

Figure 2 is a diagrammatic cross-section of a reactor canal with storage facilities in which it is convenient to expose the lube oil-olefin mixture to gamma radiation from spent fuel elements.

Figure 3 shows a fuel assembly similar to those used in the Materials Testing Reactor (MTR) operated for the Atomic Energy Commission, at The Materials Testing Reactor, National Reactor Testing Station, Idaho.

Figure 4 is a cross-section on the line 4—4 of Figure 3 with fuel plates in place.

Figure 5 shows a partial plan view of a storage rack for spent fuel elements located in the reactor canal of Figure 2.

Figure 6 is a decay curve for single spent MTR fuel element versus an assembly of eight spent fuel elements arranged as in Figure 5.

Referring to Figure 1, reactor 10 comprises a central tank 12 closed near the upper end by a lead partition 14 including a lead plug 16. Partition 14 provides a shielded operating room 18 above reactor fuel core 20 which is controlled by means of control rod 22 from room 18.

Tank 12 below partition 14 is filled with a suitable moderator 24 which may be a lube oil-olefin mixture, in one embodiment of the invention, or any conventional moderator. Tank 12 is enclosed by a compartment 26 containing a suitable reflector which may also comprise a lube oil-olefin mixture, but graphite balls or other reflector type material may be used in this compartment. Reflector compartment 26 is encased in and completely surrounded by a concrete shield 28.

In one embodiment of the invention, compartment 26 and tank 12 (below partition 14) are filled with a lube oil-olefin mixture and conduits 30 and 32 are connected with the lower and upper ends of compartment 26 for introducing and removing oil, respectively, to and from compartment 26. Control of oil flow is effected by remote operation of valves 34 and 36. Valve 38 and conduit 40 provide oil circulation means through tank 12 around fuel core 20. Conduits 30 and 32 are connected to a lube oil-olefin mixture storage tank 42 by means of valved conduits 44 and 46. A conduit 48 provided with a pump 50 circulates oil mixture from the storage tank through the reactor compartments and back to tank 42. Reaction products containing lube oil of improved characteristics may be drawn off as desired through valved conduit 52.

A fluid turbine operated electrical generator 31, having leads 33, is connected by fluid conduits 35 and 37 to an indirect heat exchanger 39 which in turn is connected with line 32 by means of conduits 41 and 43 so as to provide heat exchange with the hot oil mixture and fluid power for the turbine of generator 31. It is also feasible to supply fluid power from heat exchange with tanks 12 or 26.

Figure 2 shows an elevation partially in section of a canal 54 positioned below a reactor for storage of spent fuel assemblies 56 in racks 58. Fuel elements are delivered to the canal by means of a chute 60 which connects (by means not shown) with fuel core 20. A crane 62 which is operated from a tunnel adjacent wall 63, is provided for transporting fuel elements in the 16 to 18 feet of water in canal 54.

Figure 3 shows the shell of an MTR type fuel assembly. The shell 64 is made of aluminum plate with two flat sides 66 and 67 and two curved sides 68 and 69 (shown in Figure 4). End sections 71 and 72 are riveted and welded to the fuel section 73 to provide a cylindrical upper terminal 74 adapted to fit holes in grid (not shown) in fuel core 20 and to provide a square terminal section 77 to fit in holes 78 of a lower grid (not shown). A spring 79 is provided on cylindrical terminal 74 to compensate for vibrations arising during the operation of the reactor.

Referring to Figure 4, the fuel plates 81 are shown brazed to grooves provided in straight sides 66 and 67 of the shell 64. The curvature and spacing of the fuel plates 81 are further maintained by means of aluminum cones 82 which are brazed to the plates near each end. In the fuel elements shown in Figure 4, 18 fuel plates are utilized on a 0.117 inch spacing. The fuel plates per se are made of aluminum-uranium alloy. The uranium in the alloy contains 168 grams of U^{235} per assembly. The uranium-aluminum alloy is encapsulated in an aluminum-casing to make up the fuel plates which are approximately 0.06 inch thick. When 27 of the fuel assemblies are positioned in a 3 x 9 arrangement, the reactor will produce 30,000 kw. of energy for a period of 20 days without refueling.

Figure 5 shows a plan view of a portion of a canal storage rack 58 including an array of eight fuel elements F in rectangular geometry with respect to the center line of space 84.

When the spent fuel assemblies are removed from the reactor, they have a very high gamma radiation coming mainly from the decay of fission products and when immersed in water, the gamma radiation from a single fuel element, measured at a point 10 centimeters from the fuel element, shows an initial gamma intensity of 3×10^6 r.

per hour and decays to 10^5 r. per hour in approximately 120 days. This is illustrated in Figure 6 which is self-explanatory. Curve I is the decay diagram for spent fuel elements stored in the canal as shown in Figure 5 in a rectangular array made up of 8 spent fuel elements. On the center line of space 84, the initial gamma intensity is 10^7 r. per hour which decays in about 20 days to 3×10^6 per hour. At that time, another group of spent fuel elements is removed from the reactor and upon replacing the original spent fuel elements, the intensity is again raised to 10^7 r. per hour so that gamma intensity in the range 10^7 to 3×10^6 r. per hour is readily available on a continuous basis during the normal operation of a reactor such as the MTR.

Results of a specific example of the invention are summarized in the table. The SAE 10 lubricating oil base stock treated was prepared from a Mid-Continent crude. After an original flashing, de-salting and de-emulsification of the raw crude stock, the material is charged to a topping still wherein gasoline and light oils are removed therefrom. The residue is vacuum distilled to remove heavy gas oil and the light lubricating oils of SAE 10 viscosity and SAE 20 viscosity grades.

A portion of the SAE 10 grade base stock so prepared amounting to 137 grams was placed in a high pressure radiation container, which was a high pressure bomb, catalogue No. 41-2180 of American Instrument Company, Micro Reaction Vessel, and 38 grams of ethylene was added. The container was lowered through the water in the canal at the MTR facility into the center of an array of spent fuel elements as shown in Figure 5, and the contents thereof was exposed to gamma radiation of an average intensity of 5.6×10^6 for 18.02 hours, for a total dosage of 10^8 REP (Roentgen equivalent physical). The treatment was effected at ambient temperature, which was about 80° F.

Table

Sample.....	1	2	3	4
10 Stock.....	Unirradiated.	Irradiated.	Irradiated, Centrifuged Liquid.	Irradiated Centrifuged Jelly.
Specific Gravity.....	0.8645	.8692	.8645	
Viscosity:				
100° F.....	99.88	384.5	130.7	
210° F.....	39.38	45.17	42.68	
Index.....	100	<0	121	
Color NPA.....	1		1½	
Drop Point.....				168° F.

Sample 1 is a sample of the untreated oil, while sample 2 is a sample of the same oil after irradiation. Samples 3 and 4 were derived from sample 2 by centrifugal separation. The liquid portion (sample 3) was an excellent lubricating oil having a much improved viscosity index. The other portion was a soft wax or jelly resembling petroleum useful as a grease for lubrication of bearings and the like.

In accordance with the present invention, the treatment of the lube oil-olefin mixture with ionizing radiation is generally effected in the temperature range from -120 to 700° F. Lower temperatures are applicable but temperatures over 900° F. are not preferred since degradation of lube oil is appreciable unless the residence in the reactor or during exposure to ionizing radiation is maintained for only a short time. Temperatures of 60 to 500° F. are usually preferred.

In producing the olefin-lube oil reaction products of the invention, the olefin reacts with the oil to produce new products. To recover the modified oil of increased viscosity index, solid particles are removed from the oil by centrifugation, filtration or other means.

In accordance with the present invention, the lube oil-olefin mixture is exposed to ionizing radiation generally at a rate of 10^3 to 10^9 REP per hour, preferably 10^5 to

10⁸ REP per hour, although higher or lower rates can be employed.

It will be apparent to those skilled in the art that variations and modifications of the invention can be made from a study of the foregoing disclosure. Such variations and modifications are believed to be clearly within the spirit and scope of the invention.

We claim:

1. A process for treating a petroleum lubricating oil which comprises admixing said oil with an olefin having 2 to 5 carbon atoms in a ratio of 1-2000 parts of said olefin per 100 parts of said oil, and exposing the said admixture to a total dosage of ionizing radiation of 5×10^6 to 5×10^9 REP units, and thereby forming an oil having a higher viscosity index than the original lubricating oil, the said oil having a higher viscosity index being a product of reaction of said petroleum lubricating oil with said olefin.

2. A product of the reaction of a petroleum lubricating oil with an olefin having 2 to 5 carbon atoms in the presence of a dosage of ionizing radiation of 5×10^6 to 5×10^9 REP units, there being employed in the reaction mixture 1-2000 parts of said olefin per 100 parts of said oil.

3. A product of the reaction of a petroleum lubricating oil with an olefin having 2 to 5 carbon atoms in the presence of a dosage of gamma radiation of 5×10^6 to 5×10^9 REP units, there being employed in the reaction mixture 1-2000 parts of said olefin per 100 parts of said oil.

4. A product of the reaction of a petroleum lubricating oil with ethylene in the presence of a dosage of ionizing radiation of 5×10^6 to 5×10^9 REP units, there being employed in the reaction mixture 1-2000 parts of said ethylene per 100 parts of said oil.

5. A process of claim 1 wherein said olefin is ethylene.

6. A process of claim 1 wherein the temperature of the oil is maintained within the range from -120 to 700° F.

7. A process of claim 1 wherein the total dosage is in the range from 10^7 to 10^9 REP units.

8. A process for treating a petroleum lubricating oil which comprises admixing said oil with an olefin having 2 to 5 carbon atoms in a ratio of 1-2000 parts of said olefin per 100 parts of said oil, and exposing the said admixture to a total dosage of gamma radiation of 5×10^6 to 5×10^9 REP units, and thereby forming an oil having a higher viscosity index than the original lubricating oil, the said oil having a higher viscosity index being a product of reaction of said petroleum lubricating oil with said olefin.

9. A process of claim 8 wherein the total dosage is from 10^7 to 10^9 REP units.

10. A process of claim 7 wherein said olefin is ethylene.

11. A process of claim 9 wherein said olefin is ethylene.

12. A process of claim 1 wherein said ionizing radiation is received by said admixture while being employed as a moderator coolant in an atomic reactor, and after receiving a desired dosage of ionizing radiation, said admixture is removed from the zone of radiation of said reactor, said removed admixture being replaced by

another quantity of such admixture as moderator coolant.

13. A process for reacting ethylene with a lubricating oil which comprises exposing an admixture of 1-2000 parts of said ethylene per 100 parts of said oil to a source of ionizing radiation comprising fission products from an atomic reactor, the total dosage of exposure of said admixture to said ionizing irradiation being in the range from 5×10^6 to 5×10^9 REP units.

14. A process of claim 13 wherein the temperature of said reaction is in the range from 60 to 500° F.

15. A process for treating a petroleum lubricating oil which comprises admixing said oil with an olefin having 2 to 5 carbon atoms in a ratio of 1-2000 parts of said olefin per 100 parts of said oil, and exposing the said admixture to a total dosage of ionizing radiation of 5×10^6 to 5×10^9 REP units, separating any solids from the reaction product, and recovering an oily material having a higher viscosity index than said original petroleum lubricating oil, the said oil having a higher viscosity index being a product of reaction of said petroleum lubricating oil with said olefin.

16. A process for reacting an olefin having 2 to 5 carbon atoms with a lubricating oil which comprises exposing an admixture of 1-2000 parts of said olefin per 100 parts of said oil to a source of ionizing radiation, the total dosage of exposure of said admixture to said ionizing irradiation being in the range from 5×10^6 to 5×10^9 REP units.

17. A process for treating a petroleum lubricating oil which comprises admixing said oil with 1 to 2000 parts by weight per 100 parts of said oil of an olefin having 2 to 5 carbon atoms and exposing the said admixture to a total dosage of ionizing radiation of 5×10^6 to 5×10^9 REP units, and thereby forming an oil having a higher viscosity index than the original lubricating oil, the said oil having a higher viscosity index being a product of reaction of said petroleum lubricating oil with said olefin.

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