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(54) **Fuel compositions.**

(57) A gasoline composition for spark ignition internal combustion engines having a RON of at least 80 and containing 1 to 6% v/v of dimethyl carbonate as an octane No. improving additive.

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Fuel compositions

This invention relates to fuel compositions and in particular to gasoline compositions for use in spark ignition internal combustion engines.

Such gasoline compositions comprise a mixture of hydrocarbons and other additives. The composition is required to vaporise over a range of temperatures to give satisfactory hot and cold starting characteristics and efficient engine operation. To this end the compositions generally have initial and final boiling points within the range 25-35°C and 200-220°C respectively. The gasoline composition is often varied with the time of year and/or region to allow for variations in the average ambient temperature. The distillation and vapour pressure characteristics of typical gasolines are as follows.

		Cold weather formulation	Hot weather formulation
initial boiling point	ASTM D-86	28°C	32°C
10% boiling point		42	48-49
50% boiling point		94-100	99-104
90% boiling point		164-168	166-172
final boiling point		207-210	209-213
Reid vapour pressure (ASTM D-323)		85-86 kPa	67-68 kPa

For economic reasons the compositions generally contain as much butane as is consistent with obtaining satisfactory vapour pressure and boiling range characteristics.

Spark ignition internal combustion engines, such as those used in automobiles, generally have, in the interests of efficiency, a relatively high compression ratio. For such engines a fuel having a high octane rating is required: the Research Octane Number (RON) is normally above 80, more usually above 85, and in most cases above 90: indeed compositions having a RON in the range 95-100 are widely used for premium grade fuels. In order to achieve the desired octane rating, additives are gnerally incorporated into the composition: lead compounds, e.g. lead tetraethyl, are the most widely used octane rating improvers. However environmental considerations make the use of highly leaded gasolines undesirable: the discharge of exhaust gases from internal combustion engines into the atmosphere causes pollution and allegedly present health hazards. These exhaust gases contain not only lead compounds which themselves give rise to pollution and alleged health hazards but also various nitrogen oxides which may also be objectionable. To reduce the nitrogen oxides emission it is often desirable to employ catalytic converters in the exhaust system to convert the nitrogen oxides to less objectionable materials: however the catalysts in such converters are often poisoned by lead compounds in the exhaust gases.

For these environmental reasons the maximum amount of lead that can be incorporated into gasoline is often restricted: in many countries it is restricted to no more than 0.4 g Pb/litre and in some countries to no more than 0.15 Pb/litre. Indeed unleaded gasoline is used in some countries.

Alternative octane improving additives that have been employed include ethers and alcohols such as methyl t-butyl ether (MTBE) and t-butanol alone or in admixture with other alcohols such as ethanol. The blending RON of these additives varies to some extent on the base gasoline used in the RON determination and on the amount of additive employed: typical blending RON are as follows:

t-butanol	~ 108
ethanol	110-120
MTBE	115-133

While measurements indicate that methanol has a high blending RON, the benefit observed in normal automobile usage is markedly less than that predicted from its blending RON.

While MTBE has a high blending RON, it has a relatively low boiling point (55°C) and so its use as an octane improver has the disadvantage that the amount of butane that can be included in the gasoline composition is reduced. Methanol suffers from the same disadvantage.

We have found that di-alkyl carbonates can be used to improve the octane rating of gasoline without the aforementioned disadvantages. Di-alkyl carbonates have been proposed as gasoline additives in US-A-2331386 which quoted a blending octane number of 96-98 for di-ethyl carbonate (DEC) when used in gasolines of octane number about 74 to 79. Our measurements conducted on gasolines having higher RON indicate that DEC has a blending RON of the order of 110-112 and that, surprisingly, di-methyl carbonate (DMC) has a much higher blending RON, of the order of 120-130 or more.

Compared with DEC and other di-alkyl carbonates, e.g. di- n-propyl carbonate (DPC) and di-n-butyl carbonate (DBC), and with other lead-free octane improving additives such as alcohols, e.g. methanol, ethanol, t-butanol, and MTBE, DMC has a number of disadvantages. Inter alia it has a significantly higher specific gravity and a much lower net calorific value (i.e. the heat of combustion excluding the heat liberated by condensation of the water vapour formed during combustion, since in internal combustion engines this water vapour is not condensed but is emitted with the exhaust gases).

However, the unexpectedly high blending RON of DMC outweighs these disadvantages and renders DMC particularly useful as an octane improving agent.

Accordingly the present invention provides a gasoline composition having a RON of at least 80 and comprising gasoline hydrocarbons and from 1 to 6% by volume, based on the volume of the composition, of DMC.

Another disadvantage of octane improving additives such as alcohols is their water miscibility. The use of water miscible additives presents storage problems, particularly the use of gasoline storage tanks having water providing a base level. Not only may water miscible additives tend to be leached from the gasoline into the aqueous phase upon storage over a water base, but also their presence in the gasoline may increase the solubility of water in the gasoline.

The specific gravity, net calorific value, and water solubility of DMC in relation to other di-alkyl carbonates and other gasoline additives are listed in the following table.

Additive	Specific gravity	Net calorific value MJ/kg	Water solubility g/100 ml
DMC	1.07	14.5	11.8
DEC	0.98	21.2	1.9
DPC	0.94	~ 25 *	0.2
DBC	0.93	~ 28 *	≲ 0.02
methanol	0.79	19.8	∞
ethanol	0.79	27.1	∞
t-butanol	0.79	32.8	∞
MTBE	0.75	34.9	~ 5
gasoline	0.73 - 0.76	42.7 - 43 5	-

* estimated

Although DMC has an appreciable solubility in water, and this solubility is significantly greater than that of other di-alkyl carbonates, the partition coefficient of DMC between the gasoline and aqueous phases is strongly in favour of dissolution in the gasoline phase. Furthermore, despite the greater solubility of DMC in water, the increase in solubility of water in gasoline resulting from incorporation of DMC into gasoline appears to be significantly less than that given by the incorporation of DEC into gasoline.

DMC can readily be produced from feedstocks other than crude oil and so its use enables a greater amount of fuel to be obtained from a given quantity of crude oil.

Thus DMC can be made from methanol by reaction with carbon monoxide and oxygen over a suitable catalyst, e.g. copper chloride: for example see Ind. Eng. Chem. Prod. Res. Dev. (1980) 19, 396-403. It may also be made by reacting ethylene oxide with carbon dioxide to produce ethylene carbonate which is then reacted with methanol to produce DMC and ethylene glycol: for example see US-A-3642858 and 3803201.

The compositions of the present invention may contain other additives e.g. viscosity modifiers, gum suppressants and other octane improvers, e.g. other di-alkyl carbonates, alcohols or ethers, such as t-butanol and MTBE, and lead compounds such as lead tetraalkyls e.g. lead tetraethyl and lead tetramethyl. However, for environmental reasons mentioned hereinbefore, the lead content is preferably not more than 0.4, preferably not more than 0.15, g Pb/litre. In particular we prefer that the gasoline composition is substantially lead free.

Di-alkyl carbonates that can be used in combination with DMC (boiling point 90°C) are those di-alkyl carbonates of the formula R^1-O-R^2 in which R^1 and R^2 are alkyl radicals which may be the same or different and in which the total number of carbon atoms in the alkyl groups R^1 and R^2 is from 3 to 8. Examples of such di-alkyl carbonates include:

	Approx. boiling point (°C)
di-ethyl carbonate (DEC)	127
ethyl methyl carbonate	109
di-n-propyl carbonate (DPC)	166
di-iso-propyl carbonate	147
di-n-butyl carbonate (DBC)	208
di-iso-butyl carbonate	190

Each of the alkyl groups R^1 and R^2 preferably contains less than 5 carbon atoms.

Since di-alkyl carbonates, and DMC in particular, have a poor calorific value when compared to hydrocarbons, the total amount of di-alkyl (including di-methyl) carbonates is preferably below 10% by volume of the gasoline composition, while the amount of DMC employed, whether alone or in admixture with other di-alkyl carbonates and/or other octane improvers is between 1 and 6% by volume of the gasoline composition. The use of higher proportions of di-alkyl carbonates, and DMC in particular, would not only give less energy per litre of the composition but also would necessitate modification of carburettor or fuel injector settings to allow for the different fuel/air ratio required for the combustion of di-alkyl carbonates relative to that required for hydrocarbons. The incorporation of DMC into gasoline has no unexpected adverse effect upon the distillation characteristics or the Reid vapour pressure of the composition.

Preferably the amount of DMC employed is 3 to 5% by volume. The use of such amounts of DMC generally increases the RON of unleaded gasoline or leaded gasoline containing up to 0.4 g Pb/l by about 1-2 units.

In some cases a mixture of DMC and one or more other di-alkyl carbonates may be preferable to DMC alone because the mixture has a higher calorific value, per unit volume, a range of boiling points and vapourisability, and less mutual solubility in water compare with DMC. All these properties are advantageous in gasoline compositions for spark ignition internal combustion engines. However such a mixture will of course have a lower blending RON than DMC alone.

A mixture of di-alkyl carbonates may conveniently be made, when using a di-alkyl carbonate producing process, such as those mentioned above, wherein the reactant providing the alkyl radicals is an alcohol, by using as the alcohol a mixture of alcohols. Such a mixture of alcohols may be synthesised from a synthesis gas comprising carbon monoxide and hydrogen, by the use of a suitable catalyst. Processes for making such alcohol mixtures are well known in the art.

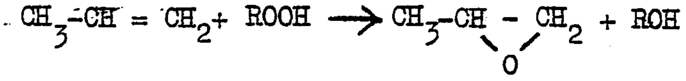
The ratio of higher alcohols (mainly C_2 to C_5) to methanol and the structure, i.e. branched or straight chain, of the alcohols higher than ethanol produced by these processes will depend on the precise catalyst and synthesis conditions, including the H_2/CO ratio employed.

As mentioned hereinbefore one convenient method for the manufacture of di-alkyl carbonates is by reaction of an alkylene oxirane e.g. ethylene or propylene oxide, with carbon dioxide to produce an alkylene carbonate which is then reacted with an alcohol to give the di-alkyl carbonate and a glycol (for example as described in US-A-3642,858 and 3,803,201). Glycols, which have a variety of uses, are often made by the hydrolysis of alkylene oxiranes. Therefore, by modification of the conventional glycol production route of hydrolysis of alkylene oxiranes by replacing that hydrolysis step with an alkylene carbonate formation step followed by reaction of the alkylene carbonate with an alcohol, a di-alkyl carbonate can be made from the alcohol in addition to the production of the glycol from the alkylene oxirane.

One method that is employed for the production of alkylene oxiranes (for subsequent hydrolysis to the corresponding glycol) involves the formation of an alkyl hydroperoxide by the reaction of an alkane with oxygen, i.e.

$$R\text{-}H + O_2 \longrightarrow R\text{-}OOH$$

followed by the reaction of the alkyl hydroperoxide with an alkene, e.g. propylene, e.g.



$$CH_3\text{-}CH = CH_2 + ROOH \longrightarrow CH_3\text{-}\underset{\diagdown\, O\, \diagup}{CH - CH_2} + ROH$$

(see for example UK-A-1,060,122 and 1,074,330). Generally the alkyl group R should be a tertiary alkyl group so that the hydroperoxide has sufficient stability for use in the reaction with the alkene. A by-product of this reaction is thus the alcohol ROH corresponding to the alkyl hydroperoxide ROOH.

The nature of the by-product alcohol ROH will of course depend on the alkane feedstock employed to make the hydroperoxide ROOH. Depending on the alkyl group R the by-product alcohol ROH can be put to a variety of uses including one or more of:

i) directly as gasoline additives,
ii) used in di-alkyl carbonate manufacture,
iii) dehydrated to the corresponding alkene which is used as part of the alkene used to make the alkylene oxirane
iv) dehydrated to the corresponding alkene which is used to make an ether
v) used as an alcohol in the reaction of an alkene and an alcohol to produce an ether.

i) Alcohols containing 4 to 8 carbon atoms, particularly t-butanol, are useful as such as gasoline additives. By the use of a mixture of alkanes, e.g. a suitable petroleum fraction, a mixture of such alcohols can be produced. Hence by integrating the abovementioned processes, a di-alkyl carbonate and one or more C_4-C_8 alcohols can be produced and used as gasoline additives, and at the same time the glycol required for other applications is produced.

ii) As mentioned hereinbefore, the alkyl group R will generally be a tertiary alkyl group. Also since, even at the theoretical 100% efficiency, only one mole of the alcohol ROH is produced for each mole of alkylene oxirane (which gives one mole of the alkylene carbonate) and two moles of alcohol are required for the reaction with the alkylene carbonate to produce the di-alkyl carbonate, some additional alcohol, e.g. methanol, obtained from another source is required, over and above that produced in the oxirane-producing reaction, for the production of the di-alkyl carbonate. Hence by using a suitable alkane feedstock to produce the alcohol ROH, and another alcohol e.g. methanol is used as the additional alcohol, a gasoline additive comprising a mixture of di-alkyl carbonates and/or a di-alkyl carbonate in which the alkyl groups differ, can be produced.

iii) Where the alcohol ROH has the same carbon skeleton as the desired glycol, the alcohol can be dehydrated to the corresponding alkene. The alkene can be recycled to the oxirane-producing reaction and used as part of the alkene employed in the production of the alkylene oxirane as described in UK-A-1,111,945. In this way the amount of alkene feedstock employed to make the glycol can be reduced, and consequently the amount of di-alkyl carbonate produced utilising a given amount of alkene feedstock is increased.

iv) The alcohol ROH can be dehydrated to an alkene which is then reacted with an alcohol to form an ether. Thus t-butanol (formed from isobutane via t-butyl hydroperoxide) dehydrates to isobutene which, on reaction with methanol, gives methyl t-butyl ether (MTBE). Again, by use of a mixture of alkanes to give a mixture of alcohols and hence a mixture of alkenes, a mixture of ethers can be produced. MTBE and similar ethers, and ether mixtures, are useful as gasoline additives.

Therefore by this route both di-alkyl carbonates and ethers which are useful as gasoline additives, can be produced in addition to a glycol.

v) The alcohol ROH can be used as part or all of the alcohol reacted with an alkene to give an ether, e.g. as described in iv) above. Some or all of the alkene may be derived from part of the alcohol ROH by dehydration as in iv) above while the remainder of the alcohol ROH is reacted, if desired in admixture with an alcohol, e.g. methanol, obtained from another source, with the alkene to give an ether, or ether mixture, suitable for use as gasoline additives.

Where a mixture of alkanes is employed, giving a mixture of hydroperoxides and hence a mixture of alcohols, the latter mixture can be fractionated so that some alcohols are subjected to one or more of the uses outlined above while others are employed in other of said uses. For example, considering uses iv) and v) above, an alcohol mixture produced from a mixture of hydroperoxides can be separated into "high" and "low" fractions: the "high" fraction can be dehydrated to the corresponding alkene or alkenes while the "low" fraction reacted with the alkene or alkene mixture obtained from the "high" fraction to give the ether or ether mixture.

Which, if any, of the aforementioned uses of the alcohol is adopted will of course depend on the nature of the feedstock employed and on the desired end product: for example in some cases it may be desired to produce a liquid fuel material containing as additives, not only at least one di-alkyl carbonate but also, at least one alcohol and at least one ether in specified proportions. By utilising a combination of uses i), iv) and/or v) and optionally ii) if necessary, in the appropriate proportions, the desired additives in their desired respective quantities can be obtained. Of course, where a mixture of alkanes is used, giving a mixture of the alcohols ROH, it may be desirable to fractionate the alcohol mixture and subject the different fractions to the different uses.

Accordingly a further aspect of the invention provides a process for the manufacture of i) a di-alkyl carbonate product consisting of at least one di-alkyl carbonate, ii) an alcohol product A consisting of at least one alcohol and/or an ether product consisting of at least one ether, and iii) a glycol product consisting of at least one glycol, comprising

a) reacting at least one alkyl hydroperoxide with an alkene feedstock B consisting of at least one alkene to produce at least one alkylene oxirane and an alcohol material C containing said alcohol product A,

b) reacting at least part of said at least one alkylene oxirane with carbon dioxide to form at least one alkylene carbonate,

c) reacting said at least one alkylene carbonate with an alcohol component D, consisting of at least one alcohol and which may contain part of said alcohol material C, to form said di-alkyl carbonate product and said glycol product,

d) separating said glycol product from said di-alkyl carbonate product and, optionally

e) dehydrating at least part of said alcohol material C to an alkene component E and etherifying said alkene component E by reaction with an alcohol component F, consisting of at least one alcohol and which may contain part of said alcohol material C, to produce said ether product.

The above process can thus upgrade an alkane feedstock into an alcohol product useful as a gasoline additive as such or as a reactant for the production of a gasoline additive, and an alkene feedstock into a glycol product and also upgrade said alcohol product or a different alcohol component into a di-alkyl carbonate product which is useful as a gasoline additive.

Accordingly a further aspect of the invention provides, in a process wherein an alkane feedstock and an alkene feedstock are upgraded to form an alcohol product and a glycol product by:

i) reacting said alkane feedstock with oxygen to form a hydroperoxide product,

ii) reacting said hydroperoxide product with said alkene feedstock to form an alkylene oxirane product and said alcohol product, and

iii) converting said alkylene oxirane product to said glycol product,

the improvement comprising also upgrading an alcohol feedstock into a gasoline additive by converting said alkylene oxirane product to said glycol product by reacting said oxirane product with carbon dioxide to form an alkylene carbonate product and reacting said alkylene carbonate product with said alcohol feedstock to form said glycol product and a di-alkyl carbonate product.

The catalysts and conditions suitable for effecting the above mentioned reactions are known and will be apparaent to those skilled in the art and so details thereof are here unnecessary.

In a preferred embodiment, isobutane is used as the alkane feedstock and the resulting hydroperoxide is reacted with propylene to give propylene oxide and t-butanol. The propylene oxide is converted to propylene carbonate which is reacted with methanol to produce propylene glycol and DMC. Alternatively ethylene is used in place of propylene thus giving ethylene glycol and DMC. The t-butanol is preferably used as such, in admixture with the DMC, as a fuel additive, or some or all of the t-butanol is dehydrated to isobutene which is etherified with methanol to give MTBE which is used in admixture with the DMC and the remainder, if any, of the t-butanol as a gasoline additive.

The invention is illustrated by the following examples.

EXAMPLE 1

Research Octane numbers (RON) of fuels were determined by the standard method (ASTM D2699) with a CFR-ASTM single-cylinder engine. Blends of 3 and 5% v/v of DMC in pure iso-octane were used. The values obtained were:

3% v/v DMC, blending RON = 130

5% v/v DMC, blending RON = 132

EXAMPLE 2

Example 1 was repeated using blends of 3 and 5% v/v of various di-alkyl carbonates in a gasoline composition comprising 80% v/v iso-octane and 20% v/v n-heptane. Motor Octane Numbers (MON) were also determined by the Standard Method (ASTM D2700). The results were as follows:

di-alkyl carbonate	% additive (v/v)	RON	MON	Average Blending No	
				RON	MON
None	0	80.0	80.0	-	-
DMC	3	81.5	80.6	131	100
	5	82.6	81.0		
DEC	3	81.0	80.7	110	103
	5	81.4	81.1		
DPC	3	80.8	80.3	110	96
	5	81.7	81.1		
DBC	3	80.1	80.0	84	81
	5	80.2	80.1		

EXAMPLE 3

Example 2 was repeated using leaded premium grade gasolines in place of the iso-octane/n-heptane mixtures. The gasolines, which differed for each di-alkyl carbonate, each contained 0.4 g Pb/l.

The results were as follows:

di-alkyl carbonate	% Additive (v/v)	RON	MON	Average Blending No	
				RON	MON
DMC	0	97.0	87.7	125	109
	3	97.9	88.3		
	5	98.3	88.8		
DEC	0	98.5	88.1	112	95
	3	98.9	88.3		
	5	99.2	88.5		
DPC	0	97.4	88.6	113	104
	3	97.8	89.0		
	5	98.3	89.1		
DBC	0	97.7	88.1	98	88
	3	97.7	88.1		
	5	97.7	88.2		

It is seen from Examples 2 and 3 that DMC gives a much higher Research Octane No improvement than other di-alkyl carbonates, both in leaded and unleaded gasolines.

EXAMPLE 4

The effectiveness of di-alkyl carbonates for lowering the surface and interfacial tension of a hydrocarbon fuel was tested as in US-A-2331386.

Precisely measured drops of a premium grade gasoline containing 3% of the di-alkyl carbonate were allowed to fall 1.2 cm on to a polished metal surface. The diameter of the resultant droplet film was measured and is expressed in the table as a percentage of that formed from the gasoline containing no di-alkyl carbonate.

Di-alkyl carbonate	Relative spread (%)
None	100
DMC	98
DEC	136
DPC	151
DBC	168

It is seen that there is reasonable agreement with the results quoted for DEC and DBC in US-A-2331386 but DMC has only a little, but negative, effect on the degree of spreading.

EXAMPLE 5

The distribution of DMC between gasoline and water was determined by shaking together at ambient temperature equal volumes of water on a commercial premium grade gasoline containing 0.4 g Pb/l to which various amounts of DMC had been added. When equilibrium had been reached, samples of each phase were analysed for DMC. The results are given in the following table.

Amount of DMC (% v/v) in		Partition coefficient
Aqueous phase	Gasoline phase	K
0.40	1.12	2.8
0.82	1.88	2.3
1.53	3.60	2.4
1.96	3.88	2 0
2.40	4.90	2.0
2.96	6.10	2.1
3.67	8.90	2.4
4.04	9.20	2.3

$$K = \frac{\text{concentration in gasoline phase}}{\text{concentration in aqueous phase}}$$

By way of comparison K for t-butanol (at a level of 3% v/v t-butanol in gasoline) is about 0.26 while that for methanol is very small.

EXAMPLE 6

The solubility of water in commercial premium grade gasolines containing 0.4 g Pb/l and various amounts of di-alkyl carbonates was determined at -7°C and at 21°C. Since the base gasoline used for the different di-alkyl carbonates differed slightly the absolute solubilities are not strictly comparable. To obtain a realistic comparison the percentage increase in solubility given by incorporation of the di-alkyl carbonate is quoted in the following table.

Temperature °C	DMC (% v/v)	DEC (% v/v)	Water concentration (% v/v)	Increase in water concentration (%)
-7	0		0.018	-
	3		0.019	6
	6		0.023	28
		0	0.019	-
		3	0.026	37
		5	0.030	58
21	0		0.039	-
	3		0.045	15
	6		0.054	38
		0	0.035	-
		3	0.046	31
		5	0.057	63

It is clear that DMC, although appreciably soluble in water does not give a large increase in the solubility of water in gasoline, whereas DEC, which has a much lower solubility than DMC in water, gives a larger increase in the solubility of water in gasoline.

1. A gasoline composition for spark ignition internal combustion engines having a Research Octane Number of at least 80 comprising gasoline hydrocarbons and 1 to 6% by volume of said composition of di-methyl carbonate.

2. A gasoline composition according to claim 1 having a lead content of no more than 0.4 g Pb/litre.

3. A gasoline composition according to claim 2 having a lead content of no more than 0.15 g Pb/litre.

4. A gasoline composition according to any one of claims 1 to 3 having a Research Octane Number of at least 90.

5. A gasoline composition according to any one of claims 1 to 4 also containing at least one other di-alkyl carbonate, the total amount of di-alkyl carbonates (including the di-methyl carbonate) being less than 10% by volume of the composition.

6. A gasoline composition according to any one of claims 1 to 5 also containing an alcohol, and/or an ether, containing a tertiary alkyl radical having from 4 to 8 carbon atoms.

7. A gasoline composition according to claim 6 wherein said alcohol is t-butanol.

8. A gasoline composition according to claim 6 wherein said ether is methyl t-butyl ether.

9. A process for the manufacture of i) a di-alkyl carbonate product consisting of at least one di-alkyl carbonate, ii) an alcohol product A consisting of at least one alcohol and/or an ether product consisting of at least one ether, and iii) a glycol product consisting of at least one glycol, comprising

a) reacting at least one alkyl hydroperoxide with an alkene feedstock B consisting of at least one alkene to produce at least one alkylene oxirane and an alcohol material C containing said alcohol product A,

b) reacting at least part of said at least one alkylene oxirane with carbon dioxide to form at least one alkylene carbonate,

c) reacting said at least one alkylene carbonate with an alcohol component D, consisting of at least one alcohol and which may contain part of said alcohol material C, to form said di-alkyl carbonate product and said glycol product,

d) dehydrating at least part of said alcohol material C to an alkene component E and etherifying said alkene component E by reaction with an alcohol component F, consisting of at least one alcohol and which may contain part of said alcohol material C, to produce said ether product.

10. A process wherein an alkane feedstock and an alkene feedstock are upgraded to form an alcohol product and a glycol product by

i) reacting said alkane feedstock with oxygen to form a hydroperoxide product,

ii) reacting said hydroperoxide product with said alkene feedstock to form an alkylene oxirane product and said alcohol product, and

iii) converting said alkylene oxirane product to said glycol product,

including the improvement comprising also upgrading an alcohol feedstock into a gasoline additive by converting said alkylene oxirane product to said glycol product by reacting said oxirane product with carbon dioxide to form an alkylene carbonate product and reacting said alkylene carbonate product with said alcohol feedstock to form said glycol product and a di-alkyl carbonate product.