

25 copolymers are segmented copolymers of ethylene and at least one other alpha-olefin monomer. At least one of the copolymer is intramolecularly heterogeneous and intermolecularly homogeneous and at least one of the copolymer, constituting at least 10% of the copolymer's chain, is a crystallizable segment. "crystallizable segment" is defined to be each segment of the copolymer chain having a number average molecular weight of at least 700 wherein the ethylene content is at least 55 wt.%. The remaining segments of the copolymer chain are herein termed the "low crystallinity segments" and are characterized by an ethylene content of not greater than about 53 wt.%. Furthermore, the molecular weight distribution of the copolymer is very narrow.

30 However, during the preparation, processing, and/or storage of these oil soluble ethylene/alpha-olefin copolymers a haze may develop in their oil concentrates or oil compositions (e.g., oil concentrates or formulations containing the ethylene-alpha-olefin copolymers). The source of this haze does not appear to be the same as that haze resulting from incompatibility of the several additives in a lubricating oil concentrate or composition (see U.S. Patent No. 3,897,353 wherein haze resulting from incompatibility is overcome in a lubricating oil additive concentrate by blending an amorphous ethylene/alpha-olefin copolymer with an n-alkyl methacrylate containing polymer having a number average molecular weight between about 30,000 and about 120,000). Rather, it is believed that this haze is due to the presence of a variety of catalysts, metal weak acid salts, etc. which are used in or result from the by-product of the polymerization, finishing process, or other steps in the manufacture or finishing of the ethylene/alpha-olefin copolymers of this invention or their oil concentrates. Thus, for example, a typical haze producing agent is calcium stearate having a particle size of from about 0.01 microns to about 15 microns, which is used in the finishing process of these ethylene-alpha-olefin copolymers useful as V.I. improvers. 35
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45 oil compositions such as oil concentrates containing these ethylene-alpha-olefin copolymer V.I. improvers also contain a haze forming amount of these metal weak acid salts. These haze forming agents are generally less than about 1 wt. % based on the total weight of the oil compositions.

It has now been surprisingly discovered that the haze of oil compositions containing these ethylene/alpha-olefin copolymer viscosity index improvers can be reduced or substantially eliminated by the addition or treatment thereof with a hydrocarbyl substituted succinic acid, a weak acid containing two dissociating moieties both of which have pKs above 2.5 (e.g., succinic acid has a pK₁ of 4.16 and a pK₂ of 5.64).

It has been discovered that the haze in lubricating oil compositions containing certain specific ethylene/alpha-olefin copolymeric V.I. improvers can be reduced or substantially eliminated by the addition of the copolymer or its oil composition such as an oil concentrate composition, which typically comprises a lubricating oil and from 0.01 to 50, preferably 3 to 30 wt. % based upon said composition, of said copolymer with a hydrocarbyl substituted succinic acid. The present invention has particular utility in reducing the hazing substance is a metal salt of a weak acid, said weak acid having a pK of more than about 3 and a pK of 4.0 to about 8 and said hazing substance has a particle size of from about 0.01 micron to about 15 microns.

acid.

25 Preferably said temperature is from 50 to 160°C.

The invention includes as an aspect the use of a hydrocarbyl succinic acid for the treatment of oil compositions containing ethylene/alpha-olefin copolymer viscosity index improving additive for reducing haze due to the inclusion in such composition of a haze forming material resulting from manufacture or finishing processes of said viscosity index improving additive, said composition defined above.

30 The ethylene-alpha-olefin copolymers are segmented copolymers of ethylene and at least one alpha-olefin monomer; each copolymer is intramolecularly heterogeneous and intermolecularly homogeneous and at least one segment of the copolymer, constituting at least 10% of the copolymer's chain, is a crystallizable segment. For the purposes of this application, the term "crystallizable segment" is defined to be a segment of the copolymer chain having a number-average molecular weight of at least 700 wherein the ethylene content is at least 55 wt.%. The remaining segments of the copolymer chain are herein termed the "non-crystallizable segments" and are characterized by an average ethylene content of not greater than about 55 wt.%. Furthermore, the molecular weight distribution (MWD) of copolymer is very narrow. It is well known that the breadth of the molecular weight distribution can be characterized by the ratios of various molecular weight averages. For example, an indication of a narrow MWD in accordance with the present invention is a ratio of weight-to-number-average molecular weight ($\bar{M}_w/\bar{M}_n$) is less than 2. Alternatively, a ratio of molecular weight to the weight-average molecular weight ($\bar{M}_z/\bar{M}_w$) of less than 1.8 typifies a narrow MWD in accordance with the present invention. It is known that a portion of the property advantages of the present invention are related to these ratios. Small weight fractions of non-crystallizable segments disproportionately influence these ratios while not significantly altering the property advantages of the copolymer on them. For instance, the presence of a small weight fraction (e.g. 2%) of low molecular weight non-crystallizable segments can depress $\bar{M}_n$, and thereby raise $\bar{M}_w/\bar{M}_n$ above 2 while maintaining $\bar{M}_z/\bar{M}_w$ less than 1.8. Therefore, in accordance with the present invention, are characterized by having at least one of $\bar{M}_w/\bar{M}_n$ less than 2 and $\bar{M}_z/\bar{M}_w$ less than 1.8. The copolymer comprises chains within which the ratio of the monomers varies along the chain length. To obtain the intramolecular compositional heterogeneity and narrow MWD, the copolymer is preferably made in a tubular reactor.

45 In accordance with the instant invention it has been discovered that the haze of an oil composition, such as a lubricating oil concentrate composition, containing a specific type of ethylene-alpha-olefin copolymer viscosity index improver can be reduced or substantially eliminated to treating said composition with a reducing effective amount of a hydrocarbyl substituted succinic acid.

55 In a preferred embodiment of the instant invention haze is reduced or substantially eliminated in a lubricating oil concentrate composition suitable for use in a lubricating oil composition comprising said oil and from about 0.01 to about 50 wt.% based on the weight of said concentrate composition.

copolymer sample. It is characterized in terms of at least one of the ratios of weight-average average molecular weight, $\bar{M}_w/\bar{M}_n$, and z-average to weight-average molecular weight, $\bar{M}_z/\bar{M}_w$ where:

$$\bar{M}_w = \frac{\sum N_i M_i^2}{\sum N_i M_i}$$

$$\bar{M}_n = \frac{\sum N_i M_i}{\sum N_i}$$

and

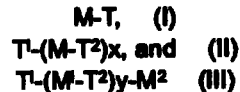
$$\bar{M}_z = \frac{\sum N_i M_i^3}{\sum N_i M_i^2}$$

wherein N_i is the number of molecules of molecular weight M_i .

d. Viscosity Index (V.I.) is the ability of a lubricating oil to accommodate increases in temperature and minimum decrease in viscosity. The greater this ability, the higher the V.I.

The instant copolymers are segmented copolymers of ethylene and at least one other monomer wherein the copolymer's chain contains at least one crystallizable segment of ethylene.

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wherein M and T are defined above, M¹ and M² can be the same or different and are each M segments, T¹ and T² can be the same or different and are each T segments, x is an integer of from 1 to 3 and y is 1 to 3.

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In structure II (x=1), the copolymer's M segment is positioned between two T segments, and the M segment can be positioned substantially in the center of the polymer chain (that is, the T¹ and T² segments have substantially the same molecular weight and the sum of the molecular weight of the T¹ and T² segments is substantially equal to the molecular weight of the M segment), although this is not essential to the practice of this invention. Preferably, the copolymer will contain only one M segment per chain. Therefore, structure I and II (x=1) are preferred.

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Preferably, the M segments and T segments of the copolymer are located along the copolymer chain such that only a limited number of the copolymer chains can associate before the steric problems associated with packing the low crystallinity T segments prevents further agglomeration. Therefore, in a preferred embodiment, the M segment is located near the center of the copolymer chain and only one M segment is in the chain.

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As will be shown below, a copolymer of the structure



(wherein M¹, M² and T are as defined above, and wherein z is an integer of at least 1) are undesirable as viscosity modifier polymers. It has been found that solutions of structure IV copolymers in oil tend to gel and the M and T portions have exactly the same composition and molecular weight as structure II copolymers (x=z=1). It is believed this poor viscosity modifier performance is due to the inability of a center M segment to sterically stabilize against association.

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The M segments of the copolymers of this invention comprise ethylene and can also comprise other alpha-olefin, e.g., containing 3 to 18 carbon atoms. The T segments comprise ethylene and other alpha-olefin, e.g., alpha-olefins containing 3 to 18 carbon atoms. The M and T segments can also comprise other polymerizable monomers, e.g., non-conjugated dienes or cyclic mono-olefins.

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Since the present invention is considered to be most preferred in the context of ethylene-propylene copolymers it will be described in detail in the context of EPM.

The ethylene-alpha-olefin copolymer in accordance with the present invention is preferably produced in a tubular reactor. When produced in a tubular reactor with monomer feed only at the tube inlet, it is at the beginning of the tubular reactor, ethylene, due to its high reactivity, will be preferentially polymerized. The concentration of monomers in solution changes along the tube in favor of propylene as the ethylene is consumed. The result, with monomer feed only at the inlet, is copolymer chains which are higher in ethylene content in the chain segments grown near the reactor inlet (as defined at the point at which the polymerization begins).

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25 as illustrated in "Polymerization of ethylene and propylene to amorphous copolymers with catalysts
oxychloride and alkyl aluminum halides"; E. Junghanns, A. Gumboldt and G. Bier; Makromol.
(12/12/62): 18-42, wherein a p-xylene/dimethylformamide solvent/non-solvent was used to
copolymer into fractions of differing intermolecular composition. Other solvent/non-solvent systems
used as hexane/2 propanol, as will be discussed in more detail below.

30 The Inter-CD of copolymer in accordance with the present invention is such that 95 wt. % of the
chains have an ethylene composition that differs from the average weight percent ethylene composition
wt. % or less. The preferred Inter-CD is about 13% or less, with the most preferred being about 10%
In comparison, Junghanns et al. found that their tubular reactor copolymer had an Inter-CD of greater
wt. %.

35 Broadly, the Intra-CD of copolymer in accordance with the present invention is such that the
portions of an individual intramolecularly heterogeneous chain, each portion comprising at least 5
percent of the chain, differ in composition from one another by at least 7 weight percent ethylene.
Otherwise indicated, this property of Intra-CD as referred to herein is based upon at least two 5 wt.
portions of copolymer chain. The Intra-CD of copolymer in accordance with the present invention is
that at least two portions of copolymer chain differ by at least 10 weight percent ethylene. Differing
40 at least 20 weight percent, as well as, of at least 40 weight percent ethylene are also considered
in accordance with the present invention.

45 The experimental procedure for determining Intra-CD is as follows. First the Inter-CD is determined as
described below, then the polymer chain is broken into fragments along its contour and the Inter-CD of the
fragments is determined. The difference in the two results is due to Intra-CD as can be seen in the following
example below.

Consider a heterogeneous sample polymer containing 30 monomer units. It consists of three
designated A, B, C.

50 A EEEEPEEEPEEEPPPEPPPEPPPEPPPPPPPP
 B EEEEPEEEPEEEPPPEEEPPPEPPPEEEPPP
 C EEPEEEPEEEPEEEPEEEPPPEPPPEEEPPP

55 Molecule A is 36.8 wt. % ethylene, B is 46.6%, and C is 50% ethylene. The average ethylene
the mixture is 44.3%. For this sample the Inter-CD is such that the highest ethylene polymer contains
more ethylene than the average while the lowest ethylene content polymer contains 7.5% less
the average. Or, in other words, 100 weight % of the polymer is within +5.7% and -7.5% ethylene

is described in "Polymer Fractionation", M. Cantow editor, Academic 1967, p. 341 and in H. Inagaki
"Developments in Polymer Characterization", 3, 1, (1982).

For non-crystalline copolymers of ethylene and propylene, molecular weight governs insolubility
does composition in a solvent/non-solvent solution. High molecular weight polymer is less soluble
solvent mix. Also, there is a systematic correlation of molecular weight with ethylene content for the
described herein. Since ethylene polymerizes much more rapidly than propylene, high ethylene polymer
tends to be high in molecular weight. Additionally, chains rich in ethylene tend to be less soluble in
hydrocarbon/polar non-solvent mixtures than propylene-rich chains. Furthermore, for crystalline
solubility is significantly reduced. Thus, the high molecular weight, high ethylene chains are easily
on the basis of thermodynamics.

A fractionation procedure is as follows:

Unfragmented polymer is dissolved in n-hexane at 23°C to form ca a 1% solution (1 g. polymer in
hexane). Isopropyl alcohol is titrated into the solution until turbidity appears at which time the polymer
allowed to settle. The supernatant liquid is removed and the precipitate is dried by pressing between
polyethylene terephthalate film at 150°C. Ethylene content is determined by ASTM method D-3915.
is resumed and subsequent fractions are recovered and analyzed until 100% of the polymer is collected.
titrations are ideally controlled to produce fractions of 5-10% by weight of the original polymer, covering
the extremes of composition.

To demonstrate the breadth of the distribution, the data are plotted as % ethylene versus the
weight of polymer as defined by the sum of half the weight % of the fraction of that composition plus
weight % of the previously collected fractions.

Another portion of the original polymer is broken into fragments. A suitable method for doing this is
thermal degradation according to the following procedure:

In a sealed container in a nitrogen-purged oven, a 2mm thick layer of the polymer is heated for 1 hour
at 330°C. (The time or temperature can be empirically adjusted based on the ethylene content and
weight of the polymer.) This should be adequate to reduce a 105 molecular weight polymer to fragments
5000 molecular weight. Such degradation does not substantially change the average ethylene content of the
polymer, although propylene tends to be lost on scission in preference to ethylene. This polymer is analyzed
by the same procedure as the high molecular weight precursor. Ethylene content is measured on selected
molecular weight on selected fractions.

The procedure to characterize intramolecular heterogeneity is laborious and even when performed at the
absolute optimum, does not show how the segments of the chain are connected. In fact it is not possible with
current technology, to determine the polymer structure without recourse to the synthesis conditions.
knowledge of the synthesis conditions, the structure can be defined as follows.

Ethylene, propylene or high alpha-olefin polymerization with transition metal catalysts can be

in the reacting medium. Thus, the ratio of $[E]/[P]$ will decrease as the monomers are consumed. will the composition in the polymer equal that in the reacting medium.

If the amount of monomer that has reacted at a given time in a batch reactor or at a given point in a reactor can be determined, it is possible through equation (1), to determine the instantaneous composition being formed at a given point along the polymer chain. Demonstration of narrow MWD and increase in the tube proves the compositional distribution is intramolecular. The amount of polymer formed is determined in either of two ways. Samples of the polymerizing solution may be collected, with quenching to terminate the reaction at various points along the reactor, and the amount of polymer evaluated. Alternatively, if the polymerization is run adiabatically and the heat of polymerization is known, the amount of monomer converted may be calculated from the reactor temperature profile.

Finally, if the average composition of the polymer is measured at a series of locations along the reactor at various times in the batch polymerization case, it is possible to calculate the instantaneous composition of the polymer being made. This technique does not require knowledge of R_1 and R_2 or the heat of polymerization but it does require access to the polymer synthesis step.

All of these methods have been employed with consistent results.

For the purpose of this patent, R_1 and R_2 thus simply serve to characterize the polymer composition of the polymerization conditions. By defining R_1 and R_2 , we are able to specify the intramolecular compositional distribution. In the examples shown below where VC1₄ and ethylaluminum sesquichloride are used in hexane as solvent, $R_1 = 1.8 \exp(+500/RT_k)$ and $R_2 = 3.2 \exp(-1500/RT_k)$. Where "R" is the gas constant (cal/deg-mole) and "T_k" is degrees Kelvin. For reference, at 20°C $R_1 = 9.7$, $R_2 = 0.02$.

The R_1 and R_2 given above predict the correct final average polymer composition. If the R_1 and R_2 expressions (2) are someday proven to be inaccurate the polymer intramolecular compositional distribution will remain as defined herein in terms of the polymerization conditions but may have to be modified on different composition scales. There is little likelihood that they are in error by more than a few percent, however.

Ethylene content is measured by ASTM-D3900 for ethylene-propylene copolymers between 0 and 85 wt.% ethylene. Above 85% ASTM-D2238 can be used to obtain methyl group concentrations which can be converted to percent ethylene in an unambiguous manner for ethylene-propylene copolymers. When comonomers other than propylene are employed no ASTM tests covering a wide range of ethylene contents are available. Proton and carbon-13 nuclear magnetic resonance spectroscopy can be employed to determine the composition of such polymers. These are absolute techniques requiring no calibration when operated properly. All nuclei of a given element contribute equally to the spectra. For ranges not covered by the ASTM methods for ethylene-propylene copolymers, these nuclear magnetic resonance methods can also be used.

Molecular weight and molecular weight distribution are measured using a Waters 150C gel permeation chromatography equipped with a Chromatix KMX-6 (LDC-Milton Roy, Riviera Beach, Fla.) light scattering photometer. The system is used at 135°C with 1,2,4 trichlorobenzene as mobile phase.

25 norbornenes such as 5-methylene-2-norbornene (MNB), 5-ethylidene-2-norbornene (ENB), 2-norbornene, 5-isopropylidene-2-norbornene, 5-(4-cyclopentenyl)-2-norbornene; 5-cyclopropylidene-2-norbornene.

Of the non-conjugated dienes typically used to prepare these copolymers, dienes containing of the double bonds in a strained ring are preferred. The most preferred diene is 5-ethylidene-2-norbornene (ENB). The amount of diene (wt. basis) in the copolymer could be from about 0% to 20% with 0% preferred. The most preferred range is 0% to 10%.

30 As already noted, the most preferred copolymer in accordance with the present invention is polyethylene. The average ethylene content of the copolymer could be as low as about 20% on a weight basis. A more preferred minimum is about 25%. A more preferred minimum is about 30%. The maximum ethylene content could be about 90% on a weight basis. The preferred maximum is about 85%, with the most preferred maximum is about 80%. Preferably, the copolymers of this invention intended for use as viscosity modifiers contain from about 35 to 75 wt.% ethylene, and more preferably from about 50 to 70 wt.% ethylene.

35 The molecular weight of copolymer made in accordance with the present invention can vary over a wide range. It is believed that the weight-average molecular weight could be as low as about 2,000. The minimum is about 10,000. The most preferred minimum is about 20,000. It is believed that the maximum weight-average molecular weight could be as high as about 12,000,000. The preferred maximum is about 1,000,000. The most preferred maximum is about 750,000. An especially preferred range of weight-average molecular weight for copolymers intended for use as V.M. polymer is from 50,000 to 500,000.

40 The copolymers of this invention will also be generally characterized by a Mooney viscosity (at 125°C) of from about 1 to 100, preferably from about 10 to 70, and more preferably from about 10 to 50. The copolymers will also be characterized by a thickening efficiency ("TE") of from about 0.4 to 5.0, preferably from about 1.0 to 4.0, most preferably from about 1.4 to 3.8.

45 Another feature of copolymer of the present invention is that the molecular weight distribution is very narrow, as characterized by at least one of a ratio of $\overline{M}_w/\overline{M}_n$ of less than 2 and a ratio of $\overline{M}_w/\overline{M}_z$ of less than 1.8. As relates to EPM and EPDM, a typical advantage of such copolymers having narrow molecular weight distribution is resistance to shear degradation. Particularly for oil additive applications, the preferred copolymer has a $\overline{M}_w/\overline{M}_z$ ratio less than about 1.5, with less than about 1.25 being most preferred. The preferred $\overline{M}_w/\overline{M}_n$ is less than 1.5, with less than about 1.2 being most preferred.

50 The copolymers of the instant invention may be produced by polymerization of a reaction mixture of catalyst, ethylene and at least one additional alpha-olefin monomer, wherein the amounts of monomers, preferably ethylene, is varied during the course of the polymerization in a controlled manner as hereinafter described. Solution polymerizations are preferred.

Any known solvent for the reaction mixture that is effective for the purpose can be used in solution polymerizations in accordance with the present invention. For example, suitable solvents

- 25 (a) the catalyst system produces essentially one active catalyst species,
(b) the reaction mixture is essentially free of chain transfer agents, and
(c) the polymer chains are essentially all initiated simultaneously, which is at the same time reactor or at the same point along the length of the tube for a tubular reactor.

To prepare copolymer structures II and III above (and, optionally, to prepare copolymer structures I, additional solvent and reactants (e.g., at least one of the ethylene, alpha-olefin and diene) will be added along the length of a tubular reactor or during the course of polymerization in a batch reactor, or in stages of stirred reactors in series in a controlled manner (as will be hereinafter described) to produce copolymers of this invention. However, it is necessary to add essentially all of the catalyst at the start of the tube or at the onset of batch reactor operation to meet the requirement that essentially all polymer chains be initiated simultaneously.

35 Accordingly, polymerization in accordance with the present invention are carried out:

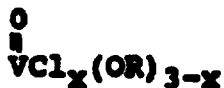
- (a) in at least one mix-free reactor,
(b) using a catalyst system that produces essentially one active catalyst species,
(c) using at least one reaction mixture which is essentially transfer agent-free, and
(d) in such a manner and under conditions sufficient to initiate propagation of essentially all polymer chains simultaneously.

40 Since the tubular reactor is the preferred reactor system for carrying out polymerizations in accordance with the present invention, the following illustrative descriptions are drawn to that system, but will be applicable to other reactor systems as will readily occur to the artisan having the benefit of the present disclosure.

In practicing polymerization processes in accordance with the present invention, use is made of at least one tubular reactor. Thus, in its simplest form, such a process would make use of but a single reactor. However, as would readily occur to the artisan having the benefit of the present disclosure, a series of reactors could be used with multiple monomer feed to vary intramolecular composition as described below.

The composition of the catalyst used to produce alpha-olefin copolymers has a profound effect on the copolymer product properties such as compositional dispersity and MWD. The catalyst utilized in processes in accordance with the present invention should be such as to yield essentially one active catalyst species in the reaction mixture. More specifically, it should yield one primary active catalyst species which provides for substantially all of the polymerization reaction. Additional active catalyst species could be present as much as 35% (weight) of the total copolymer. Preferably, they should account for about 10% of the total copolymer. Thus, the essentially one active species should provide for at least 85% of the total copolymer produced, preferably for at least 90% thereof. The extent to which a catalyst species controls polymerization can be readily determined using the below-described techniques for characterizing catalyst according to the number of active catalyst species.

55 Techniques for characterizing catalyst according to the number of active catalyst species are described below.



where $x = 0-3$ and $R =$ a hydrocarbon radical;

30

VC1_4 ;

$\text{VO}(\text{AcAc})_2$,

where $\text{AcAc} =$ acetyl acetate which may or may not be alkyl-substituted (e.g., to C_8 alkyl);

$\text{V}(\text{AcAc})_3$;

$\text{V}(\text{dicarbonyl moiety})_3$;

35

$\text{VOCl}_x(\text{AcAc})_{3-x}$,

where $x = 1$ or 2 ;

$\text{V}(\text{dicarbonyl moiety})_3\text{Cl}$; and

$\text{VC1}_3.n\text{B}$,

where $n=2-3$, $\text{B} =$ Lewis base capable of making hydrocarbon-soluble complexes with V

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tetrahydrofuran, 2-methyl-tetrahydrofuran and dimethyl pyridine, and the dicarbonyl moiety is a dicarbonyl compound of the formula:

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In formula (1) above, each R (which can be the same or different) preferably represents a C_1 to C_8 alicyclic or aromatic hydrocarbon radical such as ethyl (Et), phenyl, isopropyl, butyl, propyl, n-butyl, hexyl, cyclohexyl, octyl, naphthyl, etc. R' preferably represents an alkylene divalent radical between two carbons (e.g., $-\text{CH}_2-$, $-\text{C}_2\text{H}_4-$, etc.).

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Nonlimiting illustrative examples of formula (1) compounds are vanadyl trihalides, alkoxy halides such as VOCl_3 , $\text{VOCl}_2(\text{OBu})$ where $\text{Bu} =$ butyl, and $\text{VO}(\text{OC}_2\text{H}_5)_3$. The most preferred vanadium compounds are VC1_4 , VOC1_3 , and $\text{VOC1}_2(\text{OR})$.

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As already noted, the co-catalyst is preferably organo-aluminum compound. In terms of chemical structure, these compounds could be as follows:

25 known that certain combinations of vanadium and aluminum compounds that can comprise the catalyst can cause branching and gelation during the polymerization for polymers containing high levels of vinyl. To prevent this from happening Lewis bases such as ammonia, tetrahydrofuran, pyridine, tetrahydrothiophene, etc., can be added to the polymerization system using techniques well known to those skilled in the art.

Chain transfer agents for the Ziegler-catalyzed polymerization of alpha-olefins are well known. Illustrated, by way of example, by hydrogen or diethyl zinc for the production of EPM and EPDM. These are very commonly used to control the molecular weight of EPM and EPDM produced in continuous stirred tank reactors. For the essentially single active species Ziegler catalyst systems used in accordance with the present invention, addition of chain transfer agents to a CFSTR reduces the polymer molecular weight but does not affect the molecular weight distribution. On the other hand, chain transfer reactions during the polymerization in accordance with the present invention broaden polymer molecular weight distribution. Inter-CD. Thus the presence of chain transfer agents in the reaction mixture should be minimized or eliminated altogether. Although difficult to generalize for all possible reactions, the amount of chain transfer agent should be limited to those amounts that provide copolymer product in accordance with the desired limits as regards MWD and compositional dispersity. It is believed that the maximum amount of chain transfer agent that can be present in the reaction mixture could be as high as about 0.2 mol/mol of transition metal, e.g., vanadium, provided that the resulting copolymer product is in accordance with the desired limits as regards molecular weight and compositional dispersity. Even in the absence of added chain transfer agent, chain transfer reactions can occur because propylene and the organo-aluminum cocatalyst can also act as chain transfer agents. Among the organo-aluminum compounds that in combination with the vanadium compound yield the most active catalyst species, the organo-aluminum compound that gives the highest copolymer molecular weight and highest catalyst activity should be chosen. Furthermore, if the Al/V ratio has an effect on the molecular weight of the copolymer product, that Al/V should be used which gives the highest molecular weight also for the highest catalyst activity. Chain transfer with propylene can best be limited by avoiding excessively elevated temperatures during the polymerization as described below.

50 Molecular weight distribution and Inter-CD are also broadened by catalyst deactivation during the polymerization which leads to termination of growing chains. It is well known that the vanadium Ziegler catalysts used in accordance with the present invention are subject to such deactivation reactions which depend to an extent upon the composition of the catalyst. Although the relationship between catalyst lifetime and catalyst system composition is not known at present, for any given catalyst, deactivation can be reduced by using the shortest residence time and lowest temperature in the reactor that will produce the desired monomer conversions.

Polymerizations in accordance with the present invention should be conducted in such a manner that conditions sufficient to initiate propagation of essentially all copolymer chains simultaneously are provided.

25 reactor inlets should be high enough to provide complete, rapid chain initiation at the start of the polymerization reaction. The length of time the reaction mixture spends at high temperature must be short enough to minimize the amount of undesirable chain transfer and catalyst deactivation reactions.

Temperature control of the reaction mixture is complicated somewhat by the fact that the polymerization reaction generates large quantities of heat. This problem is, preferably, taken care of by using preheated solvent to the reactor to absorb the heat of polymerization. With this technique, the reactor is operated adiabatically and the temperature is allowed to increase during the course of polymerization. As an alternative, prechilled, heat can be removed from the reaction mixture, for example, by a heat exchanger surrounding a portion of the reactor or by well-known autorefrigeration techniques in the case of batch reactors or stirred reactors in series.

35 If adiabatic reactor operation is used, the inlet temperature of the reactor feed could be about 100°C to 150°C. It is believed that the outlet temperature of the reaction mixture could be as high as 160°C. The preferred maximum outlet temperature is about 70°C. The most preferred maximum is about 50°C. In the absence of reactor cooling, such as by a cooling jacket, to remove the heat of polymerization, the temperature is determined (for a mid-range ethylene content EP copolymer and a solvent with heat capacity similar to that of the reaction mixture) that the temperature of the reaction mixture will increase from reactor inlet to outlet by about 13°C for each percent of copolymer in the reaction mixture (weight of copolymer per weight of solvent).

40 Having the benefit of the above disclosure, it would be well within the skill of the art to determine the operating temperature conditions for making copolymer in accordance with the present invention. For example, assume an adiabatic reactor and an outlet temperature of 35°C are desired for a reaction mixture containing 5% copolymer. The reaction mixture will increase in temperature by about 13°C for each weight percent of copolymer or $5 \text{ wt}\% \times 13^\circ\text{C}/\text{wt}\% = 65^\circ\text{C}$. To maintain an outlet temperature of 35°C, it will thus be necessary that the feed inlet temperature be $35^\circ\text{C} - 65^\circ\text{C} = -30^\circ\text{C}$. In the instance that external cooling is used to absorb the heat of polymerization, the feed inlet temperature could be higher with the other temperature constraints above otherwise being applicable.

45 Because of heat removal and reactor temperature limitations, the preferred maximum copolymer concentration at the reactor outlet is 25 wt./100 wt. diluent. The most preferred maximum concentration is 10 wt./100 wt. There is no lower limit to concentration due to reactor operability, but for economic reasons it is preferred to have a copolymer concentration of at least 2 wt/100 wt. Most preferred is a concentration of at least 3 wt/100 wt.

50 The rate of flow of the reaction mixture through the reactors should be high enough to provide rapid mixing of the reactants in the radial direction and minimize mixing in the axial direction. Good radial mixing is not only to both the intra- and inter-CD of the copolymer chains but also to minimize radial temperature gradients due to the heat generated by the polymerization reaction. Radial temperature gradients in the case of segment polymers will tend to broaden the molecular weight distribution of the copolymer.

ethylene glycol monoethyl ether tribromoacetate, butyl perchlorocrotonate and methyl perchlorocrotonate. By practicing processes in accordance with the present invention, alpha-olefin copolymers of narrow MWD can be made by direct polymerization. Although narrow MWD copolymers can be made by other known techniques, such as by fractionation or mechanical degradation, these techniques are generally to be impractical to the extent of being unsuitable for commercial-scale operation. As regards EPR made in accordance with the present invention, the products have good shear stability and (intramolecular CD) excellent low temperature properties which make them especially suitable for applications.

It is preferred that the Intra-CD of the copolymer is such that at least two portions of an intramolecularly heterogeneous chain, each portion comprising at least 5 weight percent of said comonomer, differ in composition from one another by at least 5 weight percent ethylene. The Intra-CD can be such that two portions of copolymer chain differ by at least 10 weight percent ethylene. Differences of at least 15 percent, as well as, 40 weight percent ethylene are also considered to be in accordance with the present invention.

It is also preferred that the Inter-CD of the copolymer is such that 95 wt.% of the copolymer has an ethylene composition that differs from the copolymer average weight percent ethylene composition by 10 wt.% or less. The preferred Inter-CD is about 13% or less, with the most preferred being about 5%.

OIL SOLUBLE HYDROCARBYL SUBSTITUTED SUCCINIC ACID

In accordance with the practice of this invention, the heavy oil concentrate compositions or oils are treated with the oil-soluble hydrocarbyl substituted succinic acid. The hydrocarbyl moiety of the succinic acid may be alkenyl or alkyl. The hydrocarbyl moiety contains at least a sufficiently long carbon chain to make the hydrocarbyl substituted succinic acid oil soluble. Thus, the hydrocarbyl moiety contains at least about 12 carbon atoms, preferably at least about 12 carbon atoms, and more preferably at least 12 carbon atoms. The hydrocarbyl moiety contains less than about 100 carbon atoms, preferably less than about 30 carbon atoms, and more preferably less than about 20 carbon atoms. In a preferred embodiment, the hydrocarbyl substituted succinic acid is a C₁₀ to about C₂₀, preferably a C₁₂ to about C₁₈, more preferably a C₁₂ to about C₁₆, and most preferably a C₁₂ hydrocarbyl, preferably alkyl substituted succinic acid. The preferred hydrocarbyl substituted succinic acids may be represented by the general formula

wt.%, based upon the total weight of the oil composition, of said copolymer. The oil composition
25 concentrate compositions containing the ethylene-alpha-olefin copolymer V.I. Improver additive w
and can be treated according to the invention generally contain a hazing agent derived from a diss
containing material such as a metal salt of a weak organic acid. A weak organic acid has an acid n
a pK of more than about 3.8 usually a pK of 4 to 8. The hazing agent typically has a particle size
0.01 microns to about 15 microns and is present in a concentration of less than 1 wt. %, more usu
30 0.1 wt. % based on the weight of the composition. The amount of the hazing materials pres
compositions is generally dependent upon the amount of copolymer viscosity index improver
compositions contain. Generally, however, this amount is less than about 1 wt. % based on the
composition.

These metals which are found to contribute to haze include the alkaline earth metals, z
35 potassium, aluminum, vanadium, chromium, iron, manganese, cobalt, nickel, cadmium, lead,
antimony. Such metals which develop the haze can come from a variety of sources during the m
the ethylene-alpha-olefin copolymer including the catalyst, impurities developed during mechanic
of the ethylene-alpha-olefin copolymer and from dispersants used to maintain the copolymer in
suspension while stored during subsequent processing or awaiting shipping. It is generally pos
40 out those haze contributing particles which have a particle size greater than about 15 microns. A
It has been found that the haze producing impurity is difficult if not impossible to filter so that
treated according to this invention.

It is useful to carry out the process of the instant invention by first treating the ethylene
copolymer containing oil composition, e.g., an oil concentrate composition, with the hydrocarb
45 succinic acid in an amount effective to reduce or substantially eliminate the haze of said oil com
thereafter filtering out the large process debris or insoluble particulate matter. The amount o
substituted succinic acid which is effective to reduce or substantially eliminate the haze, i.e., a h
or eliminating effective amount, is any amount which is effective to reduce or preferably eliminat
said oil compositions. Generally, this amount is within the range of from about 0.001 to about 10 w
50 preferably from about 0.01 to about 1 weight percent, and more preferably from about 0.05 to abo
percent based upon the total weight of the oil composition solution.

It is to be understood that only one hydrocarbyl substituted succinic acid may be used or a
or more different hydrocarbyl substituted succinic acids may be employed.

The treatment of the haze containing ethylene-alpha-olefin copolymer oil composition is ca
55 temperature of from about room temperature to about 250°C, preferably from about 50° to abo
for a time period of about 0.1 hour up to about 20 hours, preferably from 0.5 to about 2 hours. Th
to carry out the treatment under pressure. This makes it possible to conduct the process of the i
open vessel in the presence of air or inert gas wherein the amount of haze treating agent, i.e.,

invention include those conventionally employed as crankcase lubricating oils for spark-ignited
25 sion-ignited internal combustion engines, such as automobile and truck engines, marine and
engines, and the like. Advantageous results are also achieved by employing viscosity index mod
of the present invention in base oils conventionally employed in and/or adapted for use as power
fluids such as automatic transmission fluids, tractor fluids, universal tractor fluids and hydraulic
duty hydraulic fluids, power steering fluids and the like. Gear lubricants, industrial oils, pump
30 lubricating oil compositions can also benefit from the incorporation therein of the additives of
the present invention.

Thus, the additives of the present invention may be suitably incorporated into synthetic base
alkyl esters of dicarboxylic acids, polyglycols and alcohols, polyalpha-olefins, alkyl benzenes, com
of phosphoric acids, polysilicone oils, etc.

35 Natural base oils include mineral lubricating oils which may vary widely as to their crude
whether paraffinic, naphthenic, mixed, paraffinic-naphthenic, and the like; as well as to their fo
distillation range, straight run or cracked, hydrofined, solvent extracted and the like.

More specifically, the natural lubricating oil base stocks which can be used in the compo
invention may be straight mineral lubricating oil or distillates derived from paraffinic, naphthenic
40 mixed base crudes, or, if desired, various blends oils may be employed as well as residuals, part
from which asphaltic constituents have been removed. The oils may be refined by conventional m
acid, alkali, and/or clay or other agents such as aluminum chloride, or they may be extracted
for example, by solvent extraction with solvents of the type of phenol, sulfur dioxide, furfural, d
ether, nitrobenzene, crotonaldehyde, molecular sieves, etc.

45 The lubricating oil base stock conveniently has a viscosity of typically about 2.5 to about 12, a
about 2.5 to about $9 \cdot 10^{-4} \text{ m}^2 \text{ s}^{-1}$ (cSt) at 100°C.

Thus, the additives of the present invention can be employed in a lubricating oil concentrat
or fully formulated lubricating oil composition which comprises lubricating oil, typically in a major
(I) the viscosity index improver additive, typically in a minor amount, which is effective to imp
50 viscometric properties, relative to the absence of the additive, and (II) an anti-haze effective
hydrocarbyl substituted succinic acid. Additional conventional additives selected to meet t
requirements of a selected type of lubricating oil concentrate composition or fully formulated
composition can be included as desired.

The additives of this invention, i.e., ethylene-alpha-olefin copolymer and hydrocarbyl substi
55 acid are oil-soluble, dissolvable in oil with the aid of a suitable solvent, or are stably dispersible
soluble, dissolvable, or stably dispersible as that terminology is used herein does not necessarily
the materials are soluble, dissolvable, miscible, or capable of being suspended in oil in all propor
mean, however, that the additives, for instance, are soluble or stably dispersible in oil to an ext

25 phosphosulfurized hydrocarbons and the products obtained by reaction of a phosphosulfurized
with an alkaline earth metal oxide or hydroxide, preferably in the presence of an alkylated ph
alkylphenol thioester, and also preferably in the presence of carbon dioxide. Phosphosulfurized
are prepared by reacting a suitable hydrocarbon such as a terpene, a heavy petroleum fraction
olefin polymer such as polyisobutylene, with from 5 to 30 wt. % of a sulfide of phosphorus for 1/
30 at a temperature in the range of 88° to 306°C (150° to 600°F). Neutralization of the phos
hydrocarbon may be effected in the manner taught in U.S. Patent No. 1,969,324.

Oxidation inhibitors reduce the tendency of mineral oils to deteriorate in service which d
evidenced by the products of oxidation such as sludge and varnish-like deposits on the metal s
oxidation inhibitors include alkaline earth metal salts of alkylphenolthioesters having preferably C₈ to
chains, e.g., calcium nonylphenol sulfide, barium t-octylphenyl sulfide, dioctylphenylamine, phenyl
35 amine, phosphosulfurized or sulfurized hydrocarbons, etc.

Friction modifiers serve to impart the proper friction characteristics to lubricating oil compos
automatic transmission fluids.

Representative examples of suitable friction modifiers are found in U.S. Patent No. 3,9
discloses fatty acid esters and amides; U.S. Patent No. 4,176,074 which describes molybdenu
40 of polyisobutyryl succinic anhydride-amino alkanols; U.S. Patent No. 4,105,571 which disc
esters of dimerized fatty acids; U.S. Patent No. 3,779,928 which discloses alkane phosphonic ac
Patent No. 3,778,375 which discloses reaction products of a phosphonate with an oleamide; U.
3,852,205 which discloses scarboxyalkylene hydro-carbonyl succinimide, Scarboxyalkylene
succinamic acid and mixtures thereof; U. S. Patent No. 3,879,306 which discloses N-(hydroxyalky
45 cinamic acids or succinimides; U. S. Patent No. 3,932,290 which discloses reaction products of d
phosphites and epoxides; and U. S. Patent No. 4,028,258 which discloses the alkylene ox
phosphosulfurized N-(hydroxyalkyl) alkenyl succinimides. The most preferred friction modifiers
esters, or metal salts thereof, of hydrocarbonyl substituted succinic acids or anhydrides and thiole
as described in U. S. Patent No. 4,344,853.

50 Dispersants maintain oil insolubles, resulting from oxidation during use, in suspension in
preventing sludge flocculation and precipitation or deposition on metal parts.

Pour point depressants lower the temperature at which the fluid will flow or can be p
depressants are well known. Typically of those additives which usefully optimize the low tempe
of the fluid are C₈-C₁₈ dialkylfumarate vinyl acetate copolymers, polymethacrylates, and wax
55 Foam control can be provided by an antifoamant of the polysiloxane type, e.g., silicone oil and
siloxane.

Anti-wear agents, as their name implies, reduce wear of metal parts. Representatives of cor
ti-wear agents are zinc dialkylidithiophosphate, zinc diaryldithiophosphate and magnesium sulfonate

	Anti-Wear Agents	0.001-5	.001-1
25	Friction Modifiers	0.01-5	.01-1.
	Detergents/Rust Inhibitors	.01-20	.01-3
30	Mineral Oil Base	Balance	Balance

The following examples illustrate more clearly the present invention. These examples are way of illustration and are not to be interpreted as specific limitations of the invention. In the examples otherwise indicated, all parts and percentages are on a weight basis.

35 This examples illustrates the preparation of an ethylene-propylene copolymer V.I. improver invention.

EXAMPLE 1

40 An ethylene-propylene copolymer having an ethylene content of about 56 wt. %, an $\bar{M}_w$ of 180,000, of 1.15, and a $\bar{M}_w/\bar{M}_n$ of 1.34 is prepared in a tubular reactor under the following conditions:

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25	Hexane	33,636	(74,000)
	Ethylene	2,709	(5,960)
	Propylene	3,136	(6,900)
	Total Hexane	114,091	(251,000)
30	Sidestream Feed Splits (wt.%)		
	Sidestream 1/2/3/4/5		10.3/28.8/18.9/15.9/2

35 The following example falls outside the scope of the instant invention in that the composition therein contains no hydrocarbyl substituted succinic acid. This example is presented for comparison only.

EXAMPLE 2

40 A lubricating oil concentrate is prepared containing about 6.2 wt. % of the ethylene-propylene of Example 1 by dissolving said copolymer in S-100 Neutral mineral oil. This oil concentrate is subjected to visual inspection and is found to be hazy.

The following example illustrates a composition and process of the instant invention.

EXAMPLE 3

45 There are added 0.5 gram of a S-100 Neutral oil solution of dodecyl succinic acid, containing dodecyl succinic acid, to 100 grams of the oil concentrate prepared in accordance with the Example 2. The resulting mixture is heated to 80°C. with stirring and then cooled to room temperature. The treated oil concentrate is subjected to visual inspection and is found to have less haze than the oil of Example 2.

55 In summary the preceding examples, which teach the product and process of the invention, have demonstrated that haze reduction of ethylene copolymer viscosity index improver containing oil is readily realized when such compositions are treated according to the process of this invention. The haze is reduced but these compositions remain visually improved in haze reduction for periods of time met in the shelf life required for such oil compositions.

As earlier noted the oil additive concentrate or compositions are contemplated to be admixed with additives such as zinc dihydrocarbyl dithiophosphate, and other conventional additives may also

- characterised by treating said composition for from 0.1 to 20 hours at a temperature of from 250°C with from 0.001 to 10 weight percent, based on the total weight of the composition, of a hydrocarbyl succinic acid.
2. The process of claim 1 wherein said temperature is from 50 to 160°C.
 3. The process according to claim 1 or claim 2 wherein said copolymer (II) has an inherent viscosity of from 0.1 to 0.5 dl/g in a solvent of from 1,2-dichloroethane to 1,2-dichlorobenzene at 30°C, and a compositional dispersity such that 95 weight % of said copolymer chains have a composition of from 50 to 55 weight % ethylene, preferably 50 to 53 weight % ethylene, different from said average ethylene composition.
 4. The process according to any of claims 1 to 3 wherein said low crystallinity segment comprises from 53 wt % ethylene, preferably 30 to 50 weight % ethylene.
 5. The process according to any of claims 1 to 4 wherein said copolymer (II) is characterized by an average molecular weight of from 2,000, preferably 20,000 to 12,000,000.
 6. The process according to any of claims 1 to 5 wherein said copolymer (II) has a MWD characterized by at least one and preferably both of a ratio of $\overline{M}_w/\overline{M}_n$ of less than 1.5, preferably less than 1.2, and a ratio of $\overline{M}_z/\overline{M}_w$ of less than 1.5, preferably less than 1.2.
 7. The process according to any of claims 1 to 6, wherein said copolymer (II) has a minimum ethylene content of 10%, preferably 20%, on a weight basis.
 8. The process according to claim 7 wherein said copolymer (II) has a total ethylene content of from 30 to 40 weight %, preferably about 35% on a weight basis.
 9. The process according to any of claims 1 to 8, wherein said copolymer (II) has a chain segment with at least one of the structures:

$$\begin{array}{ll} \text{(I)} & \text{M-T} \\ \text{(II)} & \text{T}^1 - (\text{M} - \text{T}^2)_x \\ \text{(III)} & \text{T}^1 - (\text{M}^1 - \text{T}^2)_y - \text{M}^2 \end{array}$$

wherein x and y are each integers of 1 to 3, M comprises said crystallizable segment, T comprises said low crystallinity segment, M¹ and M² are the same or different and each comprises an M segment, T¹ and T² are the same or different and each comprises a T segment.
 10. The process according to claim 9 wherein x in structure II is one.

finishing processes or said ethylene-alpha-olefin copolymer,
characterised by treating said composition for from 0.1 to 20 hours at a temperature
25 250°C with from from 0.001 to 10 weight percent, based on the total weight of the composition,
hydrocarbyl succinic acid.

2. The process of claim 1 wherein said temperature is from 50 to 160°C.
- 30 3. The process according to claim 1 or claim 2 wherein said copolymer (II) has an intermolecular
compositional dispersity such that 95 weight % of said copolymer chains have a composition
% or less, preferably 10 weight % or less, different from said average ethylene composition.
4. The process according to any of claims 1 to 3 wherein said low crystallinity segment comprises
35 to 53 wt % ethylene, preferably 30 to 50 weight % ethylene.
5. The process according to any of claims 1 to 4 wherein said copolymer (II) is characterized by an
average molecular weight of from 2,000, preferably 20,000 to 12,000,000.
- 40 6. The process according to any of claims 1 to 5 wherein said copolymer (II) has a MWD characterized
at least one and preferably both of a ratio of $\overline{M}_w/\overline{M}_n$ of less than 1.5, preferably less than 1.25,
or $\overline{M}_z/\overline{M}_w$ of less than 1.5, preferably less than 1.2.
7. The process according to any of claims 1 to 6, wherein said copolymer (II) has a minimum total
45 content of 10%, preferably 20%, on a weight basis.
8. The process according to claim 7 wherein said copolymer (II) has a total ethylene content of from
about 35% on a weight basis.
9. The process according to any of claims 1 to 8, wherein said copolymer (II) has a chain segment
50 with at least one of the structures:
$$\begin{array}{ll} \text{(I)} & \text{M-T} \\ \text{(II)} & \text{T}^1 - (\text{M} - \text{T}^2)_x \\ \text{(III)} & \text{T}^1 - (\text{M}^1 - \text{T}^2)_y - \text{M}^2 \end{array}$$

wherein x and y are each integers of 1 to 3, M comprises said crystallizable segment, T comprises
55 low crystallinity segment, M¹ and M² are the same or different and each comprises an M segment,
T¹ and T² are the same or different and each comprises a T segment.
10. The process according to claim 9 wherein x in structure II is one.



25 19. Use according to claim 17 or claim 18 wherein said hydrocarbyl succinic acid is as defined in claims 13 to 16.

20. Use according to any one of claims 17 to 19 wherein said lubricating oil composition is an oil

30 Patentansprüche

1. Verfahren zur Verringerung der Trübung in einer Schmierölzusammensetzung, bei dem die Zusammensetzung

(i) Schmieröl,

35 (ii) 0,01 bis 50 Gew.-% der Gesamtzusammensetzung eines Copolymeren von Ethylen und einem anderen α -Olefinmonomer, wobei das Copolymer intramolekular heterogene und inhomogene Copolymerketten umfaßt, die mindestens ein kristallisierbares Segment aus Methyleneinheiten und mindestens ein Ethylen- α -Olefin-Copolymersegment mit geringen Anteilen an Methyleneinheiten enthalten, wobei das mindestens eine kristallisierbare Segment mindestens 10 Gew.-% der Copolymerkette ausmacht und mindestens 57 Gew.-% Ethylen enthält und das Segment eine Kristallinität nicht mehr als 53 Gew.-% Ethylen enthält und wobei das Copolymer ein Molekulargewichtsverhältnis von M_w/M_n von weniger als 2 und ein Verhältnis von M_z/M_w von weniger als 1,8 aufweist, wobei mindestens zwei Teile einer individuellen intramolekular heterogenen Kette, von der Kette aus, die mindestens 5 Gew.-% der Kette ausmacht, sich in ihrer Zusammensetzung voneinander unterscheiden, mindestens 7 Gew.-% Ethylen unterscheiden,

45 (iii) eine trübungsbildende Menge eines öln unlöslichen trübungsbildenden Materials umfaßt, wobei das trübungsbildende Material dem Herstellungsverfahren des Ethylen- α -Olefin-Copolymeren zugegeben wird, dadurch gekennzeichnet, daß die Zusammensetzung 0,1 bis 20 Stunden bei einer Temperatur von 50 bis 250°C bezogen auf das Gesamtgewicht der Zusammensetzung mit 0,001 bis 10 Gew.-% Kohlenwasserstoffbernsteinsäure behandelt wird.

2. Verfahren nach Anspruch 1, bei dem die Temperatur 50 bis 160°C beträgt.

3. Verfahren nach Anspruch 1 oder 2, bei dem das Copolymer (ii) einen solchen intermolekularen Dispersionsgrad besitzt, daß 95 Gew.-% der Copolymerketten eine Zusammensetzung besitzen, die 15 Gew.-% oder weniger, vorzugsweise 10 Gew.-% oder weniger von der durch die Ethylenzusammensetzung verschieden ist.



25

10. Verfahren nach Anspruch 9, bei dem x in Struktur II 1 ist.

11. Verfahren nach Anspruch 9 oder 10, bei dem die T¹- und T²-Segmente im wesentlichen mittlere Molekulargewicht (Gewichtsmittel) besitzen.

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12. Verfahren nach Anspruch 11, bei dem in dem Copolymer (II) die Summe der mittleren Molekulargewichte (Gewichtsmittel) der T¹- und T²-Segmente im wesentlichen gleich dem mittleren Molekulargewicht (Gewichtsmittel) des M-Segments ist.

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13. Verfahren nach einem der Ansprüche 1 bis 12, bei dem die kohlenwasserstoffsubstituierte Bernsteinsäure eine C₁₀-C₁₀₀-kohlenwasserstoffsubstituierte Bernsteinsäure und vorzugsweise eine C₁₂-C₂₀-kohlenwasserstoffsubstituierte Bernsteinsäure ist.

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14. Verfahren nach Anspruch 13, bei dem die kohlenwasserstoffsubstituierte Bernsteinsäure eine C₁₂-C₁₆-kohlenwasserstoffsubstituierte Bernsteinsäure ist.

15. Verfahren nach Anspruch 14, bei dem der Kohlenwasserstoffsubstituent eine Alkylgruppe

16. Verfahren nach Anspruch 15, bei dem die alkylsubstituierte Bernsteinsäure Dodecylbernsteinsäure

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17. Verwendung einer Kohlenwasserstoffbernsteinsäure zur Behandlung von Schmierölzusatzungen, die viskositätsindexverbesserndes Ethylen/α-Olefin-Copolymer-Additiv enthält, für die Vermeidung von Trübungen aufgrund des Vorhandenseins in derartigen Zusammensetzungen von trübungsbildenden Materialien, das aus den Herstellungs- oder Verarbeitungsverfahren des viskositätsindexverbessernden Additivs resultiert, wobei die Zusammensetzung der Definition in den vorangehenden Ansprüchen entspricht.

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18. Verwendung nach Anspruch 17, bei der die Kohlenwasserstoffbernsteinsäure einen Kohlenstoffgehalt von mindestens 3 besitzt.

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19. Verwendung nach Anspruch 17 oder 18, bei der die Kohlenwasserstoffbernsteinsäure der Definition in einem der Ansprüche 13 bis 16 entspricht.



- de 50 à 250°C avec 0,001 à 10 % en poids, sur la base du poids total de la composition hydrocarbyl-succinique.
2. Procédé suivant la revendication 1, dans lequel ladite température va de 50 à 160°C.
 3. Procédé suivant la revendication 1 ou la revendication 2, dans lequel le copolymère (II) a une composition intermoléculaire telle que 95 % en poids desdites chaînes de copolymère ont une composition qui diffère de 15 % en poids ou moins, de préférence de 10 % en poids ou moins, de la composition moyenne en éthylène.
 4. Procédé suivant l'une quelconque des revendications 1 à 3, dans lequel le segment de faible cristallinité comprend 20 à 53 % en poids d'éthylène, de préférence 30 à 50 % en poids d'éthylène.
 5. Procédé suivant l'une quelconque des revendications 1 à 4, dans lequel le copolymère (II) est caractérisé par une moyenne pondérale du poids moléculaire de 2000, de préférence de 20 000 à 120 000.
 6. Procédé suivant l'une quelconque des revendications 1 à 5, dans lequel le copolymère (II) a une distribution de poids moléculaire DPM caractérisée par au moins l'un des rapports suivants : M_w/M_n inférieur à 1,5, de préférence inférieur à 1,25, M_z/M_p inférieur à 1,5, de préférence inférieur à 1,2.
 7. Procédé suivant l'une quelconque des revendications 1 à 6, dans lequel le copolymère (II) a une teneur minimale totale en éthylène de 10 %, de préférence de 20 % sur base pondérale.
 8. Procédé suivant la revendication 7, dans lequel le copolymère (II) a une teneur totale en éthylène supérieure à environ 35 % sur base pondérale.
 9. Procédé suivant l'une quelconque des revendications 1 à 8, dans lequel le copolymère (II) a une structure de chaîne des segments de chaîne présentant au moins l'une des structures suivantes :

$$\begin{array}{l} \text{(I)} \quad M-T \\ \text{(II)} \quad T^1 - (M - T^2)_x \\ \text{(III)} \quad T^1 - (M^1 - T^2)_y - M^2 \end{array}$$

 où x et y sont tous deux des nombres entiers de 1 à 3, M constitue ledit segment cristallisable, T constitue le segment à faible cristallinité, M¹ et M² sont identiques ou différents et comprennent chacun un segment M, et T¹ et T² sont identiques ou différents et comprennent chacun un segment T.



25

vue de réduire le trouble dû à l'inclusion dans ladite composition d'une matière créant un trouble de la production ou des opérations finales de traitement dudit additif améliorant l'indice de viscosité, la composition étant telle que définie dans l'une quelconque des revendications précédentes.

18. Utilisation suivant la revendication 17, dans laquelle l'acide hydrocarbylsuccinique a un pK_a moins égale à 3.

30

19. Utilisation suivant la revendication 17 ou la revendication 18, dans laquelle l'acide hydrocarbylsuccinique est tel que défini dans l'une quelconque des revendications 13 à 16.

20. Utilisation suivant l'une quelconque des revendications 17 à 19, dans laquelle la composition lubrifiante est un concentré à base d'huile.

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