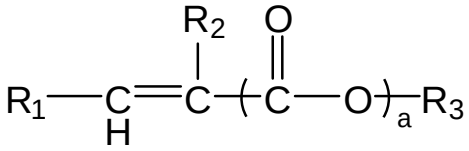
Claims

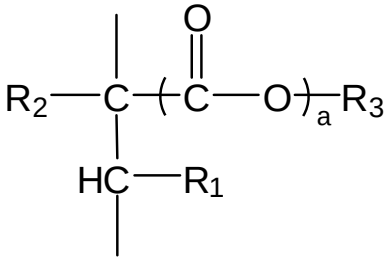
1. A gradient copolymer comprising or consisting of n polymer components, wherein the n polymer components each independently represent an addition polymer (particularly a radical addition polymer) of a monomer of formula (I),



(I),

and/or a mixture thereof, or

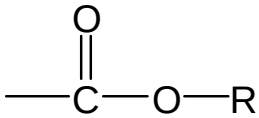
the n polymer components each independently comprise or consist essentially of one or more structural units represented by formula (I-1),



(I-1),

and the symbol n represents an integer within a closed interval [5, ∞], preferably an integer within a closed interval [8, ∞], preferably an upper limit of the integer represented by the symbol n is 20000, 10000, 5000, 1000, 500, 200, 100 or 50, and

in the formula (I) or the formula (I-1),

the group R_1 represents H or , preferably H,

the group R_2 represents H or a C_{1-4} linear or branched alkyl, preferably H or methyl,

the symbol a represents 0 or 1, preferably 1,

the group R' represents H or a group R_3, preferably the group R_3,

the group R_3 represents a C_1-C_{30} linear or branched alkyl group, preferably a C_6-C_{24} linear or branched alkyl group, more preferably a C_6-C_{20} linear alkyl group, and

where an average number of carbon atoms in a side chain of an i-th polymer component as determined according to a nuclear magnetic resonance method is expressed as X_i, with the symbol i representing an arbitrary integer from 1 to n, a following relationship holds,

$$X_1<X_2<\ldots<X_{n-1}<X_n \quad \text{(II)},$$

and there is preferably a gradual increase from X_1 to X_n, and more preferably a linear increase from X_1 to X_n.

2. The gradient copolymer according to claim 1, where a weight percentage of the i-th polymer component relative to a total weight of the n polymer components is expressed as Y_i, with the symbol i representing an arbitrary integer from 1 to n, a following relationship holds,

$$Y_1<Y_2<\ldots<Y_j>\ldots>Y_{n-1}>Y_n \quad \text{(III)},$$

in the relationship (III), the symbol j represents an integer within a closed interval [(n+1)/4, 3(n+1)/4], preferably an integer within a closed interval [(n+1)/3, 2(n+1)/3], more preferably an integer within a closed interval [2(n+1)/5, 3(n+1)/5], and $Y_1+Y_2+\ldots+Y_j+\ldots+Y_{n-1}+Y_n=100\%$,

and there is preferably a gradual increase from Y_1 to Y_j, more preferably a linear increase from Y_1 to Y_j, or there is a gradual decrease from Y_j to Y_n, and more preferably a linear decrease from Y_j to Y_n, and

more preferably, Y_i and X_i satisfy a following relationship,

$$\boldsymbol{Y_i = \mathbf{f}(X_i) = \frac{1}{\sqrt{2\pi}\sigma}\mathbf{e}^{-\frac{(X_i-\mu)^2}{2\sigma^2}}} \quad \text{(IV)},$$

in the equation (IV), the symbol μ represents an arbitrary value within an open

interval (12.5, 14.2), preferably an arbitrary value within an open interval (12.6, 13.8), and the symbol σ represents an arbitrary value within an open interval (0.5, 2).

3. The gradient copolymer according to claim 1 or 2, wherein the gradient copolymer has an average number of carbon atoms in the side chain as determined according to the nuclear magnetic resonance method within a range from 5 to 20, preferably from 11.5 to 17, preferably from 11.5. to 16.2, more preferably from 12.2 to 15.7, more preferably from 12.2 to 15.5, or a number average molecular weight Mn of the n polymer components or the gradient copolymer is each independently from 10,000 to 1,000,000, preferably from 10,000 to 500,000, more preferably from 10,000 to 100,000, or a molecular weight distribution Mw/Mn of the n polymer components or the gradient copolymer is each independently from 1.8 to 3.5, preferably from 1.9 to 3.3.

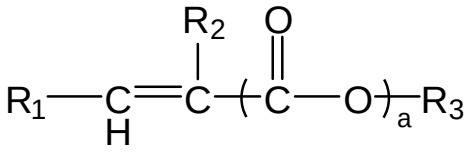
4. The gradient copolymer according to any one of claims 1 to 3, wherein the monomer of the formula (I) is one or more selected from the group consisting of C_6 linear alkyl (meth)acrylates, C_8 linear alkyl (meth)acrylates, C_{10} linear alkyl (meth)acrylates, C_{12} linear alkyl (meth)acrylates, C_{14} linear alkyl (meth)acrylates, C_{16} linear alkyl (meth)acrylates, C_{18} linear alkyl (meth)acrylates, and C_{20} linear alkyl (meth)acrylates.

5. The gradient copolymer according to any one of claims 1 to 4, wherein a proportion (by mole) of the one or more structural units represented by the formula (I-1), in which the group R_3 represents a C_{10}-C_{18} linear or branched alkyl group, relative to the total amunt of the structural units constituting each of the n polymer components is from 40% to 95%, preferably from 55% to 95%.

6. The gradient copolymer according to any one of claims 1 to 5, wherein said X_1 represents an arbitrary value within a closed interval [6.5, 12.5], preferably an arbitrary value within a closed interval [7.8, 12.0], or said X_n represents an arbitrary value within a closed interval [13.8, 19.5], preferably an arbitrary value within a closed interval [14.5, 18.2].

7. The gradient copolymer according to any one of claims 1 to 6, wherein said Y_j is within a range from 20% to 75%, preferably from 25% to 65%, or said Y_1 or Y_n is within a range from 0.01% to 20%, preferably from 0.1% to 10%.

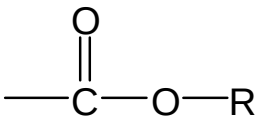
8. A method for producing a gradient copolymer, comprising steps of adding at least two monomers to a polymerization system, and conducting an addition copolymerization (particularly a radical addition copolymerization) of the at least two monomers, wherein the at least two monomers each independently represent a compound of formula (I),



(I),

and/or a mixture thereof, and

in the formula (I),

the group R_1 represents H or , preferably H,

the group R_2 represents H or a C_{1-4} linear or branched alkyl, preferably H or methyl,

the symbol a represents 0 or 1, preferably 1,

the group R' represents H or a group R_3, preferably the group R_3,

the group R_3 represents a C_1-C_{30} linear or branched alkyl group, preferably a C_6-C_{24} linear or branched alkyl group, more preferably a C_6-C_{20} linear alkyl group, and

where a starting time of adding the at least two monomers to the polymerization system is expressed as t_0, and a termination time is expressed as t_m, an addition time of the at least two monomers is defined as t ($t=t_m-t_0$), and where the addition time is divided into m equal parts, the symbol m representing an integer within a closed interval [5, ∞], preferably an integer within a closed interval [8, ∞], preferably an upper limit of the integer represented by the symbol m being 20000, 10000, 5000, 1000, 500, 200, 100 or 50, a relative ratio between the at least two monomers added to the polymerization system at any moment of time t_x, the symbol x representing an arbitrary integer from 0 to m, during the addition of monomers is such that an average number of carbon atoms in a side chain X_x of a mixture composed of the at least two monomers at the relative ratio as determined according to a nuclear magnetic resonance method satisfies a following

relationship,

$$X_0<X_1<\ldots<X_{m-1}<X_m \quad (V),$$

and there is preferably a gradual increase from X_0 to X_m, and more preferably a linear increase from X_0 to X_m.

9. The method for producing a gradient copolymer according to claim 8, where, at the termination time t_m of the addition, a sum of the cumulative amounts of the at least two monomers added to the polymerization system through the addition time t is expressed as G, and a sum of the amounts of the at least two monomers added to the polymerization system at any moment of time t_x during the addition of monomers is expressed as G_x, with the symbol x representing an arbitrary integer from 0 to m, a following relationship holds,

$$G_0/G <G_1/G <\ldots<G_j/G >\ldots>G_{m-1}/G >G_m/G \quad (VI),$$

and in the relationship (VI), the symbol j represents an integer within a closed interval [m/4, 3m/4], preferably an integer within a closed interval [m/3, 2m/3], and more preferably an integer within a closed interval [2m/5, 3m/5], and $G_0+G_1+\ldots+G_j+\ldots+G_{m-1}+G_m=G$, and

there is preferably a gradual increase from G_0/G to G_j/G, more preferably a linear increase from G_0/G to G_j/G, or there is preferably a gradual decrease from G_j/G to G_m/G, more preferably a linear decrease from G_j/G to G_m/G, and

more preferably, G_x/G and X_x satisfy a following relationship,

$$\boldsymbol{G_x/G = f(X_x) = \frac{1}{\sqrt{2\pi}\sigma} e^{-\frac{(X_x-\mu)^2}{2\sigma^2}}} \quad (VII),$$

in the equation (VII), the symbol μ represents an arbitrary value within an open interval (12.5, 14.2), preferably an arbitrary value within an open interval (12.6, 13.8), and the symbol σ represents an arbitrary value within an open interval (0.5, 2).

10. The method for producing a gradient copolymer according to claim 8 or 9, wherein the at least two monomers are two or more selected from the group consisting of a C_6 linear alkyl (meth)acrylate, a C_8 linear alkyl (meth)acrylate, a C_{10} linear alkyl (meth)acrylate, a C_{12} linear alkyl (meth)acrylate, a C_{14} linear alkyl (meth)acrylate, a C_{16} linear alkyl (meth)acrylate, and a C_{18} linear alkyl (meth)acrylate, or two or more selected

from the group consisting of a C_8 linear alkyl (meth)acrylate, a C_{10} linear alkyl (meth)acrylate, a C_{12} linear alkyl (meth)acrylate, a C_{14} linear alkyl (meth)acrylate, a C_{16} linear alkyl (meth)acrylate, a C_{18} linear alkyl (meth)acrylate and a C_{20} linear alkyl (meth)acrylate, or comprise a C_{10} linear alkyl (meth)acrylate, a C_{12} linear alkyl (meth)acrylate, a C_{14} linear alkyl (meth)acrylate, a C_{16} linear alkyl (meth)acrylate, a C_{18} linear alkyl (meth)acrylate and a C_{20} linear alkyl (meth)acrylate, preferably comprise a C_{12} linear alkyl (meth)acrylate, a C_{14} linear alkyl (meth)acrylate, a C_{16} linear alkyl (meth)acrylate, a C_{18} linear alkyl (meth)acrylate and a C_{20} linear alkyl (meth)acrylate, or comprise a C_{10} linear alkyl (meth)acrylate, a C_{14} linear alkyl (meth)acrylate, a C_{16} linear alkyl (meth)acrylate, a C_{18} linear alkyl (meth)acrylate and a C_{20} linear alkyl (meth)acrylate, or comprise a C_8 linear alkyl (meth)acrylate, a C_{14} linear alkyl (meth)acrylate, a C_{16} linear alkyl (meth)acrylate, a C_{18} linear alkyl (meth)acrylate and a C_{20} linear alkyl (meth)acrylate.

11. The method for producing a gradient copolymer according to any one of claims 8 to 10, wherein a proportion (by mole) of the compound of the formula (I), in which the group R_3 represents a C_{10}-C_{18} linear or branched alkyl group, relative to the total amount of the monomers is within a range from 40% to 95%, preferably from 55% to 95%.

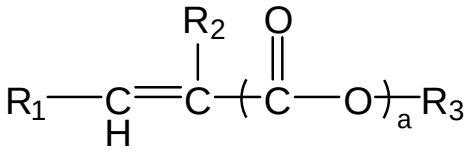
12. The method for producing a gradient copolymer according to any one of claims 8 to 11, wherein said X_0 represents an arbitrary value within a closed interval [6.5, 12.5], preferably an arbitrary value within a closed interval [7.8, 12.0], or said X_m represents an arbitrary value within a closed interval [13.8, 19.5], and preferably an arbitrary value within a closed interval [14.5, 18.2].

13. The method for producing a gradient copolymer according to any one of claims 8 to 12, wherein a ratio G_j/G is within a range from 20% to 75%, preferably from 25% to 65%, or a ratio G_0/G or a ratio G_m/G is within a range from 0.01% to 20%, preferably from 0.1% to 10%.

14. The method for producing a gradient copolymer according to any one of claims 8 to 13, wherein a reaction temperature of the copolymerization is within a range from 50 °C to 180 °C, preferably from 55 °C to 165 °C, more preferably from 60 °C to 150 °C, and a reaction time of the copolymerization is within a range from 1 hour to 24 hours, preferably from 1.5 hours to 20 hours, and an addition time t is within a range from 0.5

hours to 12 hours, preferably from 1 hour to 10 hours.

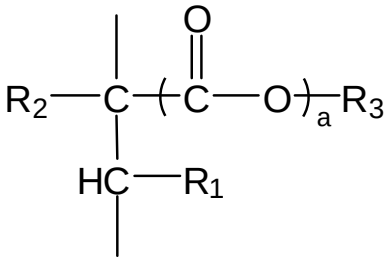
15. A method for producing a gradient copolymer, comprising a step of mixing p polymer components, wherein the p polymer components each independently represent an addition polymer (particularly a radical addition polymer) of a monomer of formula (I),



(I),

and/or a mixture thereof, or

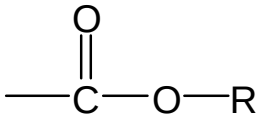
the p polymer components each independently comprise or consist essentially of one or more structural units represented by formula (I-1),



(I-1),

wherein the symbol p is an integer from 5 to 10000, preferably an integer from 8 to 5000, or an integer from 5 to 20, and

in the formula (I) or the formula (I-1),

the group R_1 represents H or , preferably H,

the group R_2 represents H or a C_{1-4} linear or branched alkyl, preferably H or methyl,

the symbol a represents 0 or 1, preferably 1,

the group R' represents H or a group R_3, preferably the group R_3,

the group R_3 represents a C_1-C_{30} linear or branched alkyl group, preferably a C_6-C_{24} linear or branched alkyl group, more preferably a C_6-C_{20} linear alkyl group,

where an average number of carbon atoms in a side chain of an i-th polymer component as determined according to a nuclear magnetic resonance method is expressed as X_i, with the symbol i representing an arbitrary integer from 1 to p, a following relationship holds,

$$X_1<X_2<\ldots<X_{p-1}<X_p \quad \text{(VIII)},$$

and there is preferably a gradual increase from X_1 to X_p, and more preferably a linear increase from X_1 to X_p.

16. The method for producing a gradient copolymer according to claim 15, where a weight percentage of the i-th polymer component relative to a total weight of the p polymer components is expressed as Y_i, with the symbol i representing an arbitrary integer from 1 to p, a following relationship holds,

$$Y_1<Y_2<\ldots<Y_j>\ldots>Y_{p-1}>Y_p \quad \text{(X)},$$

and in the equation (X), the symbol j represents an integer within a closed interval [(p+1)/4, 3(p+1)/4], preferably an integer within a closed interval [(p+1)/3, 2(p+1)/3], more preferably an integer within a closed interval [2(p+1)/5, 3(p+1)/5], and $Y_1+Y_2+\ldots+Y_j+\ldots+Y_{p-1}+Y_p=100\%$,

and there is preferably a gradual increase from Y_1 to Y_j, more preferably a linear increase from Y_1 to Y_j, or there is preferably a gradual decrease from Y_j to Y_p, more preferably a linear decrease from Y_j to Y_p, and

more preferably, Y_i and X_i satisfy a following relationship,

$$\boldsymbol{Y_i = f(X_i) = \frac{1}{\sqrt{2\pi}\sigma} e^{-\frac{(X_i-\mu)^2}{2\sigma^2}}} \quad \text{(XI)}$$

in the equation (XI), the symbol μ represents an arbitrary value within an open interval (12.5, 14.2), preferably an arbitrary value within an open interval (12.6, 13.8), and the symbol σ represents an arbitrary value within an open interval (0.5, 2).

17. The method for producing a gradient copolymer according to claim 15 or 16, wherein a proportion (by mole) of the one or more structural units represented by the formula (I-1), in which the group R_3 represents a C_{10}-C_{18} linear or branched alkyl group, relative to a total amount of the structural units constituting each of the p polymer components is within a range from 40% to 95%, preferably from 55% to 95%.

18. The method for producing a gradient copolymer according to any one of claims 15 to 17, wherein said X_1 represents an arbitrary value within a closed interval [6.5, 12.5], preferably an arbitrary value within a closed interval [7.8, 12.0], or said X_p represents an arbitrary value within a closed interval [13.8, 19.5], and preferably an arbitrary value within a closed interval [14.5, 18.2].

19. The method for producing a gradient copolymer according to any one of claims 15 to 18, wherein said Y_j is within a range from 20% to 75%, preferably from 25% to 65%, or said Y_1 or Y_p is within a range from 0.01% to 20% %, preferably from 0.1% to 10%.

20. A polymer composition comprising a gradient copolymer according to any one of claims 1 to 7, or a gradient copolymer obtained by a method for producing a gradient copolymer according to any one of claims 8 to 19.

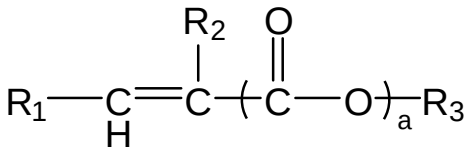
21. A pour point depressant for lubricating oils, comprising a gradient copolymer according to any one of claims 1 to 7, or a gradient copolymer obtained by a method for producing a gradient copolymer according to any one of claims 8 to 19.

22. A lubricating oil composition comprising a gradient copolymer according to any one of claims 1 to 7, or a gradient copolymer obtained by a method for producing a gradient copolymer according to any one of claims 8 to 19, or a polymer composition according to claim 20, or a pour point depressant for lubricating oils according to claim 21, and a lubricating base oil, wherein, on basis of the gradient copolymer, a content by weight of the gradient copolymer in the lubricating base oil is within a range from 0.01 wt% to 2 wt%, preferably from 0.05 wt% to 1.5 wt%, more preferably from 0.1 wt% to 1 wt%.

23. Use of a gradient copolymer according to any one of claims 1 to 7, or a gradient

copolymer obtained by a method for producing a gradient copolymer according to any one of claims 8 to 19 as a pour point depressant for lubricating oils.

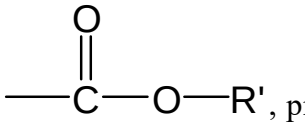
24. A method for controlling copolymerization, the copolymerization being an addition copolymerization (particularly a radical addition copolymerization) occurred in a polymerization system to which at least two monomers are added, wherein the at least two monomers each independently represent a compound of formula (I),



(I)

and/or a mixture thereof, and

in the formula (I),

the group R_1 represents H or , preferably H,

the group R_2 represents H or a C_{1-4} linear or branched alkyl, preferably H or methyl,

the symbol a represents 0 or 1, preferably 1,

the group R' represents H or a group R_3, preferably the group R_3,

the group R_3 represents a C_1-C_{30} linear or branched alkyl group, preferably a C_6-C_{24} linear or branched alkyl group, more preferably a C_6-C_{20} linear alkyl group,

where a starting time of adding the at least two monomers to the polymerization system is expressed as t_0, a termination time is expressed as t_m, an addition time of the at least two monomers is defined as t ($t=t_m-t_0$), and where the addition time is divided into m equal parts, with the symbol m representing an integer within a closed interval [5, ∞], preferably an integer within a closed interval [8, ∞], preferably an upper limit of the integer represented by the symbol m being 20000, 10000, 5000, 1000, 500, 200, 100 or 50,

the method for controlling copolymerization comprises adjusting a relative ratio between the at least two monomers added to the polymerization system at any moment of time t_x, the symbol x representing an arbitrary integer from 0 to m, during the addition of monomers, so that an average number of carbon atoms in a side chain X_x of a mixture composed of the at least two monomers at a relative ratio as determined according to a nuclear magnetic resonance method satisfies a following relationship,

$$X_0<X_1<\ldots<X_{m-1}<X_m \quad (V)$$

and there is preferably a gradual increase from X_0 to X_m, and more preferably a linear increase from X_0 to X_m.

25. The method for controlling copolymerization according to claim 24, where, at the termination time t_m of the addition, a sum of the cumulative amounts of the at least two monomers added to the polymerization system through the addition time t is expressed as G, and a sum of the amounts of the at least two monomers added to the polymerization system at any moment of time t_x during the addition of monomers is expressed as G_x, with the symbol x representing an arbitrary integer from 0 to m, a ratio G_x/G satisfies a following relationship,

$$G_0/G <G_1/G <\ldots<G_j/G >\ldots>G_{m-1}/G >G_m/G \quad (VI),$$

and in the relationship (VI), the symbol j represents an integer within a closed interval [m/4, 3m/4], preferably an integer within a closed interval [m/3, 2m/3], and more preferably an integer within a closed interval [2m/5, 3m/5], and $G_0+G_1+\ldots+G_j+\ldots+G_{m-1}+G_m=G$, and

there is preferably a gradual increase from G_0/G to G_j/G, more preferably a linear increase from G_0/G to G_j/G, or there is preferably a gradual decrease from G_j/G to G_m/G, more preferably a linear decrease from G_j/G to G_m/G, and

more preferably, G_x/G and X_x satisfy a following relationship,

$$\boldsymbol{G_x/G = f(X_x) = \frac{1}{\sqrt{2\pi}\sigma} e^{-\frac{(X_x-\mu)^2}{2\sigma^2}}} \quad (VII),$$

in the equation (VII), the symbol μ represents an arbitrary value within an open interval (12.5, 14.2), preferably an arbitrary value within an open interval (12.6, 13.8), and the symbol σ represents an arbitrary value within an open interval (0.5, 2).

26. The method for controlling copolymerization according to claim 24 or 25, wherein a proportion (by mole) of the compound of the formula (I), in which the group R_3 represents a C_{10}-C_{18} linear or branched alkyl group, relative to a total amount of the monomers is within a range from 40% to 95%, preferably from 55% to 95%.

27. The method for controlling copolymerization according to any one of claims 24 to 26, wherein said X_0 represents an arbitrary value within a closed interval [6.5, 12.5], preferably an arbitrary value within a closed interval [7.8, 12.0], or said X_m represents an arbitrary value within a closed interval [13.8, 19.5], and preferably an arbitrary value within a closed interval [14.5, 18.2].

28. The method for controlling copolymerization according to any one of claims 24 to 27, wherein a ratio G_j/G is within a range from 20% to 75%, preferably from 25% to 65%, or a ratio G_0/G or a ratio G_m/G is within a range from 0.01% to 20%, preferably from 0.1% to 10%.