Claims:

1. A process of ring formation across a carbon-carbon multiple bond, the process comprising the steps of reacting a N-alkyl-N-nitroso compound with a substrate bearing a carbon-carbon multiple bond in the presence of an aqueous base and a transition metal catalyst, wherein the N-alkyl-N-nitroso compound is generated in-situ, and the N-alkyl-N-nitroso compound is added to the substrate without being first isolated, wherein the substrate bearing a carbon-carbon multiple bond is a terminal (monosubstituted) alkene or an *exo*-methylene compound, and wherein the transition metal catalyst is a palladium catalyst.

2. A process according to claim 1 wherein the N-alkyl-N-nitroso compound is an organic solution of N-alkyl-N-nitroso urea, and wherein the N-alkyl-N-nitroso urea is added to the substrate without being first isolated in solid form.

3. A process according to claim 1 wherein the N-alkyl-N-nitroso compound is a N-methyl-N-nitroso compound (MNC).

4. A process according to claim 1 wherein the N-alkyl-N-nitroso compound is selected from the group consisting of N-methyl-N-nitroso-urea (MNU), N-methyl-N-nitroso-p-toluenesulfonamide (Diazald^{TM}), N-nitroso-dimethylurethane, nitroso-EMU and N-nitroso-β-methylaminoisobutyl methyl ketone (NMK).

5. A process according to claim 1 wherein the N-alkyl-N-nitroso compound is generated in-situ from a mixture of a N-alkylamine, water, $NaNO_2$ and an acid, before partitioning into an organic solvent to form an organic solution of N-alkyl-N-nitroso compound.

6. A process according to any of the claims 1 to 5, wherein a biphasic mixture is formed with the N-alkyl-N-nitroso compound in the organic layer.

7. A process according to any of the claims 1 through 6 wherein the N-alkyl-N-nitroso compound in liquid phase is separated from the aqueous phase in a phase separation step, before being added to the substrate bearing a carbon-carbon multiple bond.

8. A process of converting a carbon-carbon double bond to a cyclopropane ring according to any of the preceding claims.

9. A process of converting a carbon-carbon double bond to a cyclopropane ring comprising the steps of:

I) synthesis of the N-alkyl-N-nitroso compound in liquid phase,

II) separation of the organic N-alkyl-N-nitroso compound-containing liquid phase from an aqueous phase, and

III) transferring the N-alkyl-N-nitroso compound in organic liquid phase into a mixture comprising an alkene substrate, an aqueous base and a transition metal catalyst, thereby to cyclopropanate the alkene substrate, wherein the transition metal catalyst is a palladium catalyst, and wherein the alkene substrate is a terminal alkene or *exo*-methylene compound.

10. A process according to claim 5, wherein the N-alkylamine is methyl, ethyl or propyl.

11. A process according any of the claims 3 through 10 wherein the liquid phase comprises an organic solvent for the MNC that is selected from the group consisting of ethers and toluene.

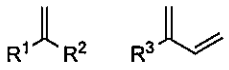
12. A process according to claim 11 wherein the ether is selected from the group consisting of tetrahydrofurane, dimethoxyethane, dioxane and dimethylisosorbide.

13. A process according to any of the claims 1 through 12 wherein the aqueous base is selected from the group consisting of alkali hydroxides.

14. A process according to any of the claims 1 through 13 wherein the catalyst is a palladium catalyst, e.g. $Pd(acac)_2$, $Pd(OAc)_2$ or $PdCl_2$.

15. A process according to any of the preceding claims, which is conducted in flow mode.

16. A process according to any of the preceding claims wherein the substrate bearing a carbon-carbon multiple bond is a compound of the formulae

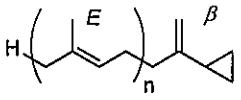


wherein R^1 and R^2 may, independently of each other, be hydrogen, alkyl, alkylidene, or aryl, which may be branched or unbranched and substituted or unsubstituted;
and R^3 may be an alkyl, alkylidene, or aryl, which may be branched or unbranched and substituted or unsubstituted.

17. A process according to claim 16 wherein the substrates are isoprenoids.

18. A Myrcene or Ocimene derivative which is mono- or bis-cyclopropanated at the terminal isoprene unit with > 70% selectivity, preferentially > 80% selectivity, preferentially > 90% selectivity.

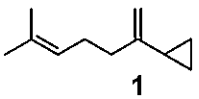
19. A compound according to the formula



IIIa

in which n = 1 or 3.

20. A compound according to claim 21 according to the formula



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21. A process according to any of claims 1 to 17 yielding a Myrcene, Ocimene, Farnesene derivative which is mono- or bis-cyclopropanated at the terminal isoprene unit with > 70% selectivity, preferentially > 80% selectivity, preferentially > 90% selectivity.