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Synthesis of Fused-Ring Indenes by Ruthenium-Catalyzed Ring-Closing Metathesis

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The preparation of new 1,2-unsymmetrically substituted fused-ring indenes from dialkenyl precursors by utilization of the ruthenium-catalyzed ring-closing metathesis is described. By analogous strategy, 1,3-substituted fused-ring indenes with medium-ring sizes have also been synthesized. For bis(allylsilyl)indene, the formation of a 1,3-fused ring appears to be prevented by ring strain and a 1,1-spiro compound is obtained instead due to rearrangement of the substituents. The rearrangement is enabled by the [1,5]-silatropic shifts operating in silyl-substituted cyclopentadienyl compounds.

Introduction

Synthetic pathways to substituted indenes continue to attract attention due to the versatility of multiply substituted indenes as building blocks for functional materials,[1,2] pharmaceuticals,[3–5] small organic molecules[6,7] and as ligand precursors for transition-metal complexes.[8–11] Recently, e.g., Halterman,[12,13] Takahashi et al.,[14] and Takai and co-workers[15] have reported efficient metal-mediated or metal-catalyzed methods for preparation of new indenyl frameworks.

Specifically, in transition-metal chemistry, group IV metallocenes incorporating substituted indenyl ligands are widely employed catalysts for stereospecific propylene polymerization providing access to polypropylene materials with a range of tacticities and material properties.[16] The microstructure and thus the properties of the polymer produced can be tailored by variations in the ligand substitution pattern. Here, unsymmetrically substituted C_1-symmetric mixed-ligand indenyl complexes with diastereotopic coordination sites often form efficient catalysts for the preparation of elastomeric polypropylenes due to alternation of isotactic and atactic stereosequences in the polymer chain produced.[17–21] In such catalysts, however, fused-ring substituents attached to the five-membered ring of the indenyl moiety have been reported in few cases only,[12,22,23,24] whereas in a number of cases beneficial effects have been related to the presence of one or two fluorenyl ligands.[25–28] In earlier work, Rabideau and co-workers have described the preparation of 1,4-dihydrofluorene by the use of metallic lithium and ammonia at low temperature, a method which is not necessarily of general use.[29,30] Towards tetrahydrofluorenes[30] similar and related synthetic routes have been explored.[12,31]

Recently, we described the simple synthesis of several allyl-substituted indenes by Zn-mediated allylation of 1- and 2-indanones in aqueous media.[32] Derivatization of the allylindenes obtained by Pt-catalyzed hydrosilylation reactions was briefly explored in the previous paper. Here, we describe the utilization of such allyl-substituted indenes in the simple preparation of new substituted fused-ring indenes by ring-closing alkene metathesis using the 2nd-generation Grubbs ruthenium catalyst 1,3-bis(2,4,6-trimethylphenyl)-2-(imidazolidinylidene)(dichlorophenylmethylene)(tricyclohexylphosphane)ruthenium [(NHC)(PCy_3)-Cl_2Ru=CHR].[33–35] The indenes prepared should prove useful as modified fluorenyl mimic ligand precursors for unsymmetric metallocene complexes enabling further tuning of the steric and electronic surroundings of the central metal in such systems. Additionally, we wished to explore the applicability of the approach for the synthesis of substituted indenes containing fused large-ring substituents, the preparation of which is likewise briefly illustrated.

Results and Discussion

Ring-Closing Metathesis of 1,2-Disubstituted Indenes

Indenes with terminally unsaturated alkyl or silyl substituents in both 1- and 2-positions were prepared in good yields by standard lithiation/alkylation or lithiation/silylation reactions of 2-allylindenes and obtained as mixtures of the expected regio- and/or stereoisomers (for details, see Exp. Sect. and Scheme 1, 2, 3, 4, and 5). Alkene isomerization towards more conjugated and thus more

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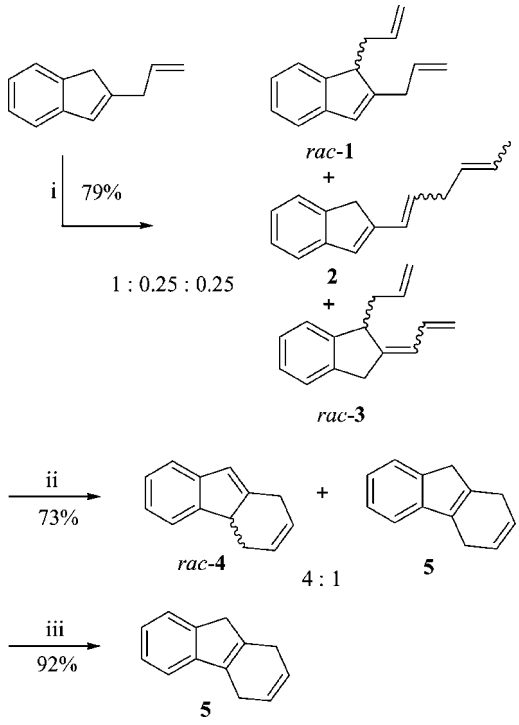
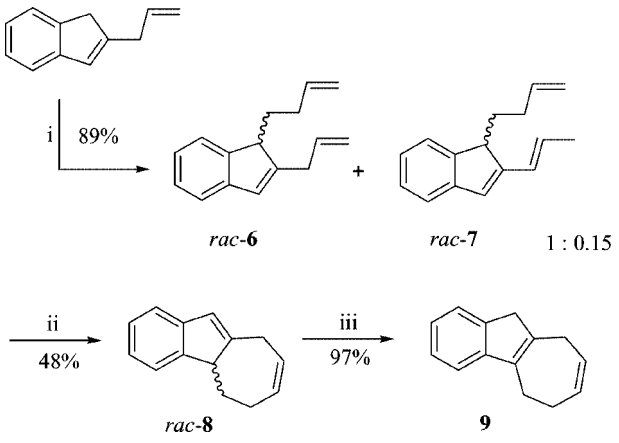
stable olefin structures were observed in some cases (**2**, *rac*-**3**, *rac*-**7** and *rac*-**11**). Because such isomerization is potentially carried further in the subsequent metathesis reactions, resulting in decreased ring sizes and in undesired side-product formation, it proved essential to carry out the alkylation/silylation sequences at low temperature in order to minimize the isomerization pathways. Unsymmetrically substituted fused-ring indenes were then prepared in moderate to good yields by treating the 1,2-disubstituted unsaturated precursors with the 2nd generation Grubbs ruthenium catalyst in dichloromethane (Scheme 1–5).

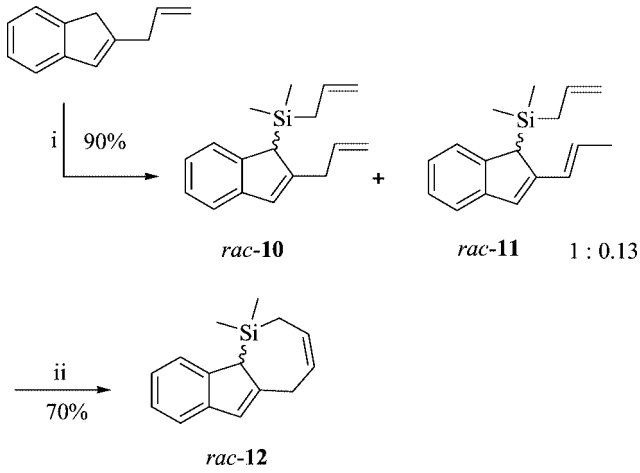


Scheme 1. (i) *n*BuLi, then allyl bromide, THF, –72 °C. (ii) Grubbs 2nd generation cat., DCM. (iii) TEA, reflux.

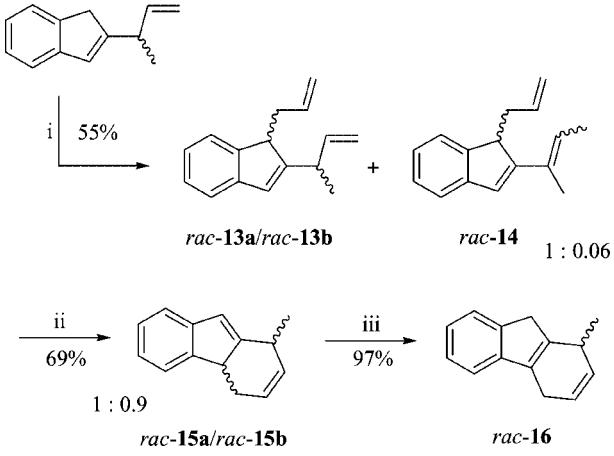


Scheme 2. (i) *n*BuLi, then 4-bromo-1-butene, THF, –72 °C. (ii) Grubbs 2nd generation cat., DCM. (iii) TEA, reflux.

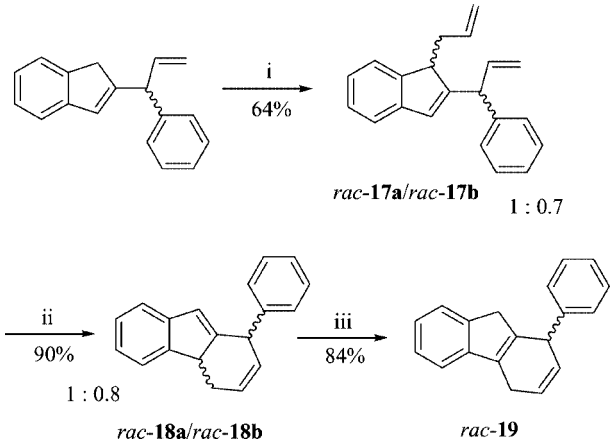
Thus, when the mixtures of *rac*-**1**/**2**/*rac*-**3**, *rac*-**6**/*rac*-**7** and *rac*-**10**/*rac*-**11** were exposed to the metathesis conditions, the fused-ring indenes *rac*-**4**/**5**, *rac*-**8** and *rac*-**12** were obtained



Scheme 3. (i) *n*BuLi, then allyl(chloro)dimethylsilane, THF, –72 °C. (ii) Grubbs 2nd generation cat., DCM.



Scheme 4. (i) *n*BuLi, then allyl bromide, THF, –72 °C. (ii) Grubbs 2nd generation cat., DCM. (iii) TEA, reflux.



Scheme 5. (i) *n*BuLi, then allyl bromide, THF, –72 °C. (ii) Grubbs 2nd generation cat., DCM. (iii) TEA, reflux.

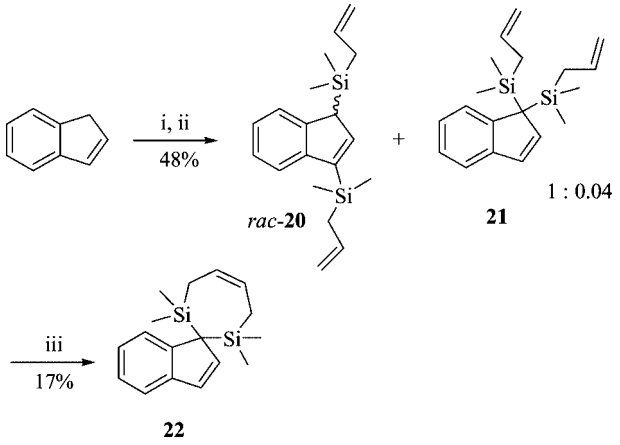
in 49–73% yields as displayed in Scheme 1, 2 and 3, and conveniently isolated by flash chromatography. In the case of the diallylindene *rac*-**1**/**2**/*rac*-**3** the ring-closing metathesis yielded a 4:1 mixture of the kinetic and thermodynamic

products *rac*-**4** and **5** which upon refluxing in triethylamine gave the thermodynamically more stable 2,3-disubstituted indene **5** as the sole product in excellent yield (Scheme 1). The ring-closing reaction of the *rac*-**6**/*rac*-**7** mixture yielded the kinetically favored 1,2-substituted indene *rac*-**8** as the only product (Scheme 2). Also this compound was converted quantitatively to the more stable thermodynamic product **9** by refluxing in triethylamine. In the case of the allylsilyl-substituted compounds *rac*-**10**/*rac*-**11**, the expected 1,2-substituted indene *rac*-**12** was obtained in good yield after purification by flash chromatography.

Analogously, when the dialkenyl indene mixtures *rac*-**13a**/*rac*-**13b**/*rac*-**14** and *rac*-**17a**/*rac*-**17b**, prepared by standard alkylations of racemic 2-(1-methylallyl)-1*H*-indene[32] and racemic 2-(1-phenylallyl)-1*H*-indene,[32] were treated with the 2nd generation Grubbs catalyst in dichloromethane, the metathesis products *rac*-**15a**/*rac*-**15b** and *rac*-**18a**/*rac*-**18b** were obtained as 1:0.9 and 1:0.8 diastereomeric mixtures, respectively, in 69 and 90% yields (Scheme 4 and Scheme 5). In both cases only the kinetic products were obtained, which again could be converted in excellent yields to the thermodynamic indenyl products *rac*-**16** and *rac*-**19** by refluxing in triethylamine.

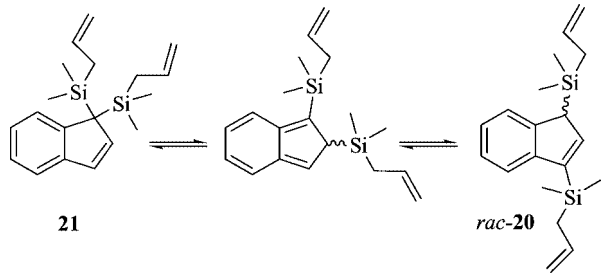
Ring-Closing Metathesis of 1,3-Disubstituted Indenes

The successful preparation of the 1,2-substituted fused-ring indenes by ring-closing metathesis motivated us to investigate a similar strategy for the preparation of analogous 1,3-substituted compounds. The 1,3-diallylsilyl-substituted indene *rac*-**20** was prepared in moderate overall yield from indene by two subsequent standard lithiation/silylation reactions and obtained, after purification by flash column chromatography, in admixture (1:0.04) with the corresponding 1,1-disubstituted product (Scheme 6).



Scheme 6. (i) *n*BuLi, THF, 0 °C, allyl(chloro)dimethylsilane. (ii) As in (i). (iii) Grubbs 2nd generation cat., DCM.

That both of these compounds are formed may be explained by reaction kinetics/thermodynamics in the lithiation/silylation sequence, where competing silylation of the indenyl 1- and 3-positions takes place. Alternatively, rearrangements may occur after the silylation reaction by the [1,5]-silatropic shifts operating in silyl-substituted indenes.[36–41] Rigby and co-workers have investigated [1,5]-silatropic shifts in disubstituted silylindenes and benzindenes showing that the initially formed, sterically crowded, 1,1-disubstituted product commonly undergoes a [1,5]-silatropic shift forming the less congested 1,3-disubstituted compound.[37] Thus, for compounds *rac*-**20** and **21**, the geminally substituted product **21** is likely to be formed first, which then, due to steric crowding, undergoes migration via isoindene to form the 1,3-substituted product (Scheme 7).

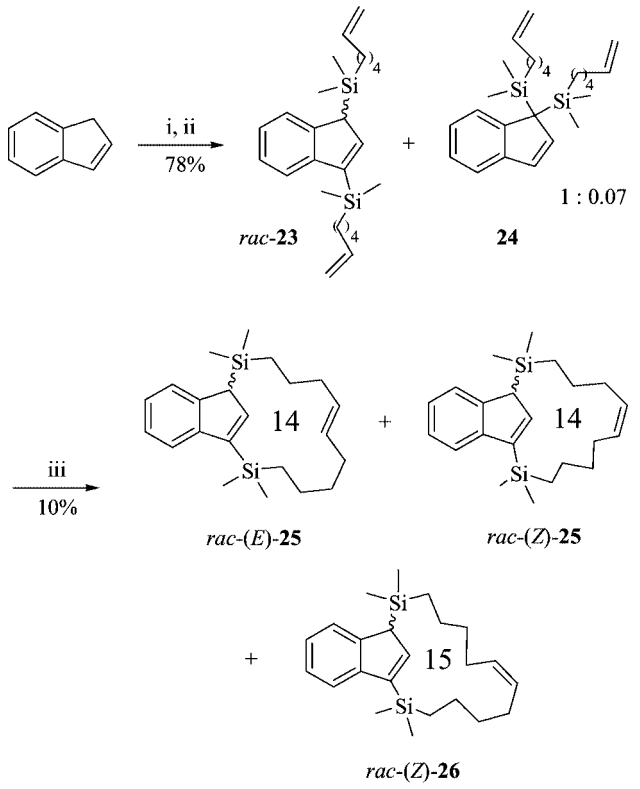


Scheme 7. [1,5]-Silatropic shifts in compounds *rac*-**20** and **21**.

Unexpectedly, when the 1:0.04 mixture of *rac*-**20** and **21** was treated with the 2nd generation Grubbs catalyst, the only metathesis product isolated was the spirocyclic compound **22**, instead of the expected 1,3-ring-bridged product, obtained in 17% isolated yield (Scheme 6). Most likely, the spiro compound **22** has been formed via [1,5]-silatropic shifts of one of the silyl groups. The ring-closing metathesis probably operates only in compound **21**; however it is prevented in the case of *rac*-**20** due to the much larger ring strain in the corresponding 1,3-bridged ring-closure product. The presumable reason for the low yield of the spirocycle **22** is that the rate of the [1,5]-silatropic shift, and thus the amount of **21** formed from *rac*-**20** at ambient temperature, is much lower than the rate of polymerization of *rac*-**20**.[42]

We investigated the factor that might govern the formation of the spirocyclic product: lowering of the activation energy by ruthenium catalyst or ring strain in the metathesis product. We prepared the analogous bis(alkenylsilyl)indene *rac*-**23**/**24** with a longer aliphatic chain that carries two hexenyl(dimethyl)silyl substituents (Scheme 8). The attempted ring-closing metathesis of this mixture produced, albeit in poor yield (10%), exclusively 1,3-bridged ring structures. Here, the rate of the [1,5]-silatropic shift is much lower at ambient temperature than the rate for ring-closing metathesis. The low yield again probably results from the polymerization of the precursor *rac*-**23**.[42]

Interestingly, the ring closure of the *rac*-**23**/**24** mixture gave, in poor yield, three different products *rac*-(*E*)-**25**, *rac*-(*Z*)-**25** and *rac*-(*Z*)-**26** in a 1:0.16:0.15 ratio. Of these, only the last mentioned have the expected 15-membered ring size. In the other two, a ring size of 14 was observed. Previously, Kinderman and co-workers have reported Ru-catalyzed isomerization reactions of terminal alkenes to more stable ones, prior to ring closing, resulting in a decreased



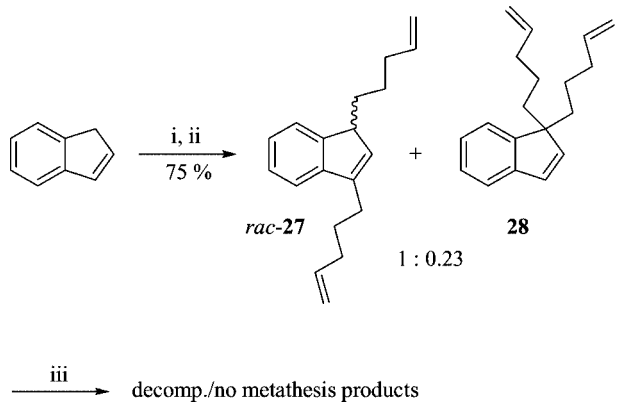
Scheme 8. (i) *n*BuLi, THF, 0 °C, 5-hexenyl(dimethyl)chlorosilane. (ii) As in (i). (iii) Grubbs 2nd generation cat., DCM.

ring size.[43] Here, the precursor *rac*-**23** must first be isomerized to form a more stable compound containing only one terminal alkene group. The isomerized compound then, upon exposure to the Grubbs catalyst and subsequent ring closure, produces a 14-membered ring instead of the expected 15-membered ring.

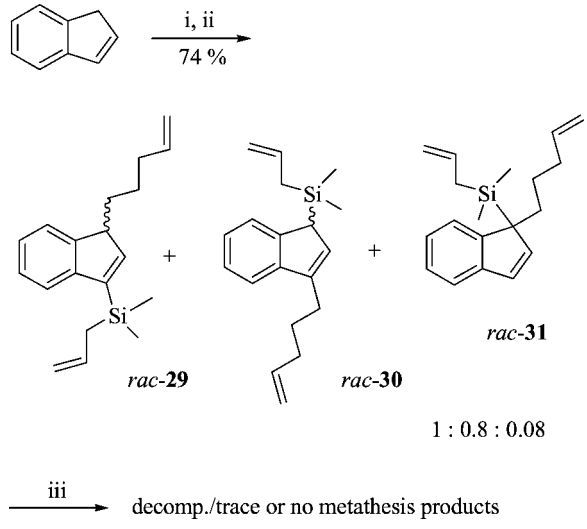
In the preceding examples reported here, for obvious steric reasons, the ring structures formed have exhibited *Z*–geometry of the double bond inside the ring system. However, when larger rings are produced, such as in the case of *rac*-(*E*/*Z*)-**25**, also (*E*)-alkenes may form as indicated by a 1:0.31 *E*/*Z*-ration for *rac*-**25** as confirmed by NMR spectroscopy.

Additionally, possibilities to prepare indenes with medium-size 1,3-ring structures containing carbon chains only were investigated. The disubstituted indene *rac*-**27** was prepared from indene in good yield by two subsequent lithiation/alkylation sequences yielding the 1,1-disubstituted analogue **28** as a side-product (Scheme 9). Exposure of the *rac*-**27**/**28** mixture to the 2nd generation Grubbs catalyst in dichloromethane gave, after 2 hours, polymerization products only, without traces of starting material or ring-closed products. Again, the ring-closing metathesis is probably prevented due to ring-strain factors.

For further insights, one of the alkenyl substituents was replaced with a silyl substituent in order to introduce a potentially migrating group to the molecule (Scheme 10). In the light of the earlier results on the ring-closing metathesis of **21** and *rac*-**23** we expected the *rac*-**29**/*rac*-**30**/*rac*-**31** mixture to yield, upon exposure to Grubbs catalyst, at least some amount of the ring-closed product, either a spirocycle or a 1,3-bridged compound. However, after the reaction at room temperature, only traces of the metathesis product were detected by MS. It can thus be concluded, that the 1,3-bridged product is too strained to be formed and only *rac*-**31** reacts under the ring-closing metathesis conditions. In this case it is likely that the activation energy for the required [1,5]-silatropic shift is too high to be overcome at ambient temperature resulting in an extremely low rate for such migration and consequently in the predominance of polymerization of *rac*-**29**/*rac*-**30**.[42]



Scheme 9. (i) *n*BuLi, THF, 0 °C, 5-bromo-1-pentene. (ii) As in (i). (iii) Grubbs 2nd generation cat., DCM.



Scheme 10. (i) *n*BuLi, THF, 0 °C, allyl(chloro)dimethylsilane. (ii) *n*BuLi, THF, 0 °C, 5-bromo-1-pentene. (iii) Grubbs 2nd generation cat., DCM, ambient temperature/reflux.

Scale-Up Experiments with 1st Generation Grubbs Catalyst

The ring-closing reactions performed here on a fairly small scale were first investigated by using the 2nd generation Grubbs catalyst which, while more expensive than the earlier reported commercially available variants, exhibits a superior performance in terms of catalyst activity and selec-

tivity. However, in terms of cost and efficiency, the scale-up of the cyclopentadienyl ligand synthesis for practical use in metallocene chemistry necessitates the preparation of these compounds in multi-gram amounts. Thus we also briefly investigated the possibilities to use the Grubbs 1st generation Ru catalyst $(PCy_3)_2Cl_2Ru{=}CHR$, bis(tricyclohexylphosphane) benzylidene, instead of the 2nd generation variant for these reactions. Accordingly, on a small scale, by analogy to Scheme 1, the ring-closing metathesis of the *rac*-**1**/**2**/*rac*-**3** mixture in refluxing dichloromethane, catalyzed by 10 mol-% of Grubbs 1st generation catalyst, yielded a mixture of *rac*-**4** and **5** in a yield comparable to that obtained from the same reaction catalyzed by the 2nd generation catalyst (see experimental section). Furthermore, our preliminary results from the scale-up experiments using the 1st generation catalyst show that the isolated yield of the ring-closing reaction can be improved from 69% to 82% when producing multigram amounts of dihydrofluorene in a single reaction. Thus, the approach described does indeed provide a practical route to new cyclopentadienyl ligand precursors.

Summary and Conclusions

To summarize, we have prepared several new 1,2-unsymmetrically substituted fused-ring indenes in moderate to excellent yields by utilizing the well-known and straightforward ring-closing metathesis reaction catalyzed by the Grubbs 2nd generation catalyst. The same method can be applied to the preparation of 1,3-substituted fused-ring indenes with medium ring sizes as well where the ring strain effects are negligible. In smaller 1,3-fused rings, however, the ring strain becomes an important factor. In the case of the 1,3-diallylsilyl-substituted indene *rac*-**20** the 1,3-substituted disilylindene rearranges to the geminal disubstituted analogue **21** via [1,5]-silatropic shifts forming the spirocyclic compound **22**.

The method described provides an attractive route to new unsymmetrically substituted cyclopentadienyl ligand precursors for transition metal complexes, the preparation of which is currently under investigation. Importantly, as demonstrated here, the ring-closing reaction has also proven amenable for scale-up by using the more economical 1st generation Grubbs catalyst without decrease in efficiency and reaction yield.

Some saturated and partially saturated fused-ring compounds, such as decahydronaphthalene, tetrahydronaphthalene and perhydrofluorene, have earlier been found to be relatively stable at high temperatures (at 648 K), required for example in the synthesis of higher alcohols.[44] These compounds could, therefore, be utilized as liquids in which the solid catalyst is suspended. Completely or partially hydroganeted 1,2-unsymmetrically substituted fused-ring indenes described here might also be stable at high temperatures and be used as suspending liquids in similar high-temperature reactors. Potential practical uses for these compounds could be found in other areas as well.

Experimental Section

General Remarks: All air- and moisture-sensitive reactions were conducted under argon using standard techniques. Commercially available solvents and reagents were used without further purification. 2-Allyl-1*H*-indene, 2-(1-methylallyl)-1*H*-indene and 2-(1-phenylallyl)-1*H*-indene were prepared as described earlier.[32] Tetrahydrofuran was distilled from sodium/benzophenone ketyl prior to use. Dichloromethane was distilled from calcium hydride prior to use. Flash chromatography was performed on silica gel 60 (40–63 μm). NMR spectra were recorded at 298 K with a Bruker Avance 600 (^{1}H NMR 600 MHz, ^{13}C NMR 150.9 MHz, ^{29}Si 119.3 MHz) or Bruker Avance 500 (^{1}H NMR 500 MHz, ^{13}C NMR 125.8 MHz). ^{1}H NMR were referenced against residual ^{1}H impurities in the solvent and ^{13}C NMR to the solvent signals. In the ^{29}Si experiments TMS was used as external reference. The NMR spectra were recorded in δ values with $CDCl_3$ as the solvent. Mass spectra were recorded with a high-resolution mass spectrometer (Fison's ZapSpec).

General Procedure 1: To a solution of monosubstituted indene in dry tetrahydrofuran (5–15 mL) at –72 °C was added *n*BuLi (2.5 M in *n*-hexane) in drops. The resulting reaction mixture was stirred at –72 °C for 4 h. To this solution was added the halide in drops at –72 °C. The reaction mixture was gradually warmed to room temperature and after a period of 1–19 h, 50 mL of water and diethyl ether were added and the formed layers were separated. The aqueous layer was further extracted with 50 mL of diethyl ether and the combined organic layers were dried with Na_2SO_4, filtered and the solvents evaporated to dryness. The crude product was purified by silica gel column chromatography (hexane as eluent) to yield the desired product/products.

General Procedure 2. Ring-Closing Metathesis (RCM) Reaction: To a solution of disubstituted indene in dry dichloromethane (2–5 mL) was added the 2nd generation $(NHC)(PCy_3)Cl_2Ru{=}CHR$ Grubbs catalyst in portions. The resulting reaction mixture was stirred at ambient temperature for a period of 30 min–24 h before the solvent was removed under reduced pressure. The residue was directly chromatographed on silica gel (hexane as eluent) and the desired product/products were obtained. The concentrations of disubstituted indenes in dichloromethane (0.18–0.25 mmol/mL, in scale-up experiment 0.36 mmol/mL) were not optimized.

General Procedure 3: The tricyclic compound/compounds were dissolved in triethylamine (4 mL) and this reaction mixture was heated to reflux for 4 h. Solvent was removed under reduced pressure and the desired compound was obtained.

General Procedure 4: To a solution of indene in dry tetrahydrofuran (15 mL) at 0 °C was added *n*BuLi (2.5 M in *n*-hexane) in drops. The resulting reaction mixture was stirred at ambient temperature for 4 h and to this solution at 0 °C was added the chlorosilane in drops. The reaction mixture was taken into room temperature and after a period of 1 h 50 mL of water and diethyl ether were added and the formed layers were separated. The aqueous layer was further extracted with 50 mL of diethyl ether and the combined organic layers were dried with Na_2SO_4, filtered and the solvents evaporated. This product was used without further purification. It was dissolved in dry tetrahydrofuran (15 mL) and to this solution was added *n*BuLi (2.5 M in *n*-hexane) in drops at 0 °C. The resulting reaction mixture was stirred at ambient temperature for 4 h and to this solution at 0 °C was added the chlorosilane in drops. The reaction mixture was taken into room temperature and after a period of 1.5 h, 50 mL of water and diethyl ether were added and the formed layers were separated. The aqueous layer was further ex-

tracted with 50 mL of diethyl ether and the combined organic layers were dried with Na_2SO_4, filtered and the solvents evaporated to dryness. The crude product was purified by silica gel column chromatography (hexane as eluent) to yield the desired product/products.

A Mixture of 1,2-Diallyl-1*H*-indene (*rac*-1), [(1*E*,4*E*)-2-Hexa-1,4-dienyl]-1*H*-indene (2) and 1-Allyl-2-prop-2-en-(*E*/*Z*)-ylidene-indane (*rac*-3): By applying the general procedure 1,2-allyl-1*H*-indene (1.0232 g, 6.5 mmol), *n*BuLi (2.6 mL, 6.5 mmol, 2.5 M in *n*-hexane) and allyl bromide (860 μL, 9.8 mmol) gave, after a 17-h reaction time, 1.0099 g (79%) of a mixture of the title compounds as a pale yellow oil. The ratio between the obtained compounds was 1:0.25:0.25. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.45–7.12 (m, 20 H, all arom. CH in *rac*-**1**, **2** and *rac*-**3**), 6.62 (s, 1 H, olefin. CH in five-ring in **2**), 6.54 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**1**), 6.43 (d, J = 16.3 Hz, 1 H, olefin. CH in chain in **2**), 5.95 (m, 8 H, 2 olefin. CH in chain in *rac*-**1**, 2 olefin. CH in chain in **2** and 4 olefin. CH in chain in *rac*-**3**), 5.51 (m, 4 H, 2 olefin. CH in chain in *rac*-**1** and 2 olefin. CH in chain in *rac*-**3**), 5.07 (m, 17 H, 4 olefin. CH_2 in chain in *rac*-**1**, olefin. CH in chain in **2** and 4 olefin. CH_2 in chain in *rac*-**3**), 3.66 (dd, J = 7.1 Hz, 4.7 Hz, 2 H, 2 aliph. CH in five-ring in *rac*-**3**), 3.46 (t, J = 6.0 Hz, 2 H, 2 aliph. CH in five-ring in *rac*-**1**), 3.36 (s, 2 H, aliph. CH_2 in five-ring in **2**), 3.35 (m, 2 H, aliph. CH_2 in chain in **2**), 3.35 (m, 2 H, 2 aliph. CH in five-ring in *rac*-**3**), 3.28–3.23 (A-part of an ABX system, dd, J_{AB} = –16.7 Hz, J_{AAX} = 6.3 Hz, 2 H, 2 aliph. CH in chain in *rac*-**1**), (The spin-system is an ABX system and the values of chemical shifts and coupling constants are approximations.), 3.26 (m, 2 H, 2 aliph. CH in five-ring in *rac*-**3**), 3.16–3.10 (a B part of an ABX system, dd, J_{AB} = –16.7 Hz, J_{AAX} = 7.3 Hz, 2 H, 2 aliph. CH in chain in *rac*-**1**)*, 2.87 (dtm, J = –14.1 Hz, 4.7 Hz, 2 H, 2 aliph. CH in chain in *rac*-**3**), 2.79 (dtm, J = –14.3 Hz, 6.0 Hz, 2 H, 2 aliph. CH in chain in *rac*-**1**), 2.50 (dtm, J = –14.3 Hz, 6.0 Hz, 2 H, 2 aliph. CH in chain in *rac*-**1**), 2.45 (dtm, J = –14.1 Hz, 7.1 Hz, 2 H, 2 aliph. CH in chain in *rac*-**3**), 1.92 (d, J = 6.7 Hz, 3 H, CH_3 in **2**) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 151.32 (2 C_q in *rac*-**1**), 149.25 (C_q in **2**), 147.47 (C_q in **2**), 146.94 (2 C_q in *rac*-**1**), 146.41 (2 C_q in *rac*-**3**), 144.75 (2 C_q in *rac*-**1**), 144.51 (C_q in **2**), 142.91 (2 C_q in *rac*-**3**), 141.26 (2 C_q in *rac*-**3**), 136.58 (olefin. CH in chain in **2**), 136.11 (2 olefin. CH in chain in *rac*-**1**), 135.69 (2 olefin. CH in chain in *rac*-**3**), 135.34 (2 olefin. CH in chain in *rac*-**3**), 135.03 (2 olefin. CH in chain in *rac*-**1**), 134.88 (2 olefin. CH in chain in *rac*-**3**), 127.95 (olefin. CH in five-ring in **2**), 127.74 (olefin. CH in chain in **2**), 127.13 (2 olefin. CH in five-ring in *rac*-**1**), 127.00 (olefin. CH in chain in **2**), 126.83, 126.18, 124.46, 124.08, 123.45, 123.38,120.78, 119.05 (12 arom. CH in **2** and *rac*-**3**), 126.73 (2 arom. CH in *rac*-**1**), 123.99 (2 arom. CH in *rac*-**1**), 123.19 (2 arom. CH in *rac*-**1**), 120.26 (2 arom. CH in *rac*-**1**), 116.68 (2 olefin. CH_2 in chain in *rac*-**1**), 116.58 (2 olefin. CH_2 in chain in *rac*-**3**), 116.42 (2 olefin. CH_2 in chain in *rac*-**1**), 115.70 (olefin. CH in chain in **2** or 2 olefin. CH_2 in *rac*-**3**), 115.67 (olefin. CH in chain in **2** or 2 olefin. CH_2 in *rac*-**3**), 50.34 (2 aliph. CH in five-ring in *rac*-**1**), 48.09 (2 aliph. CH in five-ring in *rac*-**3**), 40.68 (aliph. CH_2 in five-ring in **2**), 36.29 (2 aliph. CH_2 in chain in *rac*-**3**), 34.36 (2 aliph. CH_2 in chain in *rac*-**1**), 34.25 (2 aliph. CH_2 in chain in *rac*-**1**), 33.19 (2 aliph. CH_2 in five-ring in *rac*-**3**), 29.85 (aliph. CH_2 in chain in **2**), 18.94 (CH_3 in **2**) ppm. EIMS (70eV): calcd. $C_{15}H_{16}$ 196.1252; found 196.1248. The configuration of the diene part of the compound *rac*-**3** could not be determined due to the heavily overlapping signals in the ^{1}H NMR spectrum.

A Mixture of 4,4a-Dihydro-1*H*-fluorene (*rac*-4) and 4,9-Dihydro-1*H*-fluorene (5): By applying the General Procedure 2, a mixture of 1,2-diallyl-1*H*-indene (*rac*-**1**), [(1*E*,4*E*)-2-hexa-1,4-dienyl]-1*H*-indene (**2**) and 1-allyl-2-prop-2-en-(*E*/*Z*)-ylidene-indane (*rac*-**3**) (0.2329 g, 1.2 mmol) and Grubbs 2nd generation catalyst (0.0523 g, 0.1 mmol) gave, after a 19-h reaction time, 0.1469 g (73%) of a 4:1 mixture of the title compounds as a very pale yellow oil. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.43 (d, J = 7.5 Hz, 1 H, 1 arom. CH in **5**), 7.41 (d, J = 7.5 Hz, 2 H, 2 arom. CH in *rac*-**4**), 7.33 (d, J = 7.5 Hz, 2 H, 2 arom. CH in *rac*-**4**), 7.28 (m, 1 H, arom. CH in **5**), 7.25 (t, J = 7.5 Hz, 2 H, 2 arom. CH in *rac*-**4**), 7.23 (m, 1 H, arom. CH in **5**), 7.16 (m, 1 H, arom. CH in **5**), 7.14 (td, J = 7.5 Hz, 1.1 Hz, 2 H, 2 arom. CH in *rac*-**4**), 6.54 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**4**), 5.96 (dm, J = 9.5 Hz, 1 H, olefin. CH in six-ring in **5**), 5.90 (dm, J = 9.5 Hz, 1 H, olefin. CH in six-ring in **5**), 5.85 (m, 4 H, 4 olefin. CH in six-ring in *rac*-**4**), 3.30 (m, 4 H, 2 aliph. CH in five-ring in *rac*-**4** and alophatic CH_2 in five-ring in **5**), 3.23 (m, 4 H, 2 aliph. CH_2 in six-ring in *rac*-**4**), 3.14 (m, 4 H, 2 aliph. CH_2 in six-ring in **5**), 2.92 (m, 2 H, 2 aliph. CH in six-ring in *rac*-**4**), 1.81 (m, 2 H, 2 aliph. CH in six-ring in *rac*-**4**) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 149.12 (2 C_q in *rac*-**4**), 147.65 (2 C_q in *rac*-**4**), 145.68 (C_q in **5**), 144.97 (2 C_q in *rac*-**4**), 142.74 (C_q in **5**), 138.09 (C_q in **5**), 133.50 (C_q in **5**), 126.68 (2 arom. CH in *rac*-**4**), 126.31 (1 arom. or olefin. CH in **5**), 125.88 (2 olefin. CH in *rac*-**4**), 125.51 (2 olefin. CH in *rac*-**4**), 124.63 (1 arom. or olefin. CH in **5**), 124.33 (1 arom. or olefin. CH in **5**), 124.13 (1 arom. or olefin. CH in **5**), 123.80 (2 arom. CH in *rac*-**4**), 123.51 (1 arom. or olefin. CH in **5**), 123.23 (2 olefin. CH in five-ring in *rac*-**4**), 122.83 (2 arom. CH in *rac*-**4**), 120.45 (2 arom. CH in *rac*-**4**), 117.93 (arom. CH in **5**), 46.20 (2 aliph. CH in five-ring in *rac*-**4**), 40.44 (aliph. CH_2 in five-ring in **5**), 30.46 (2 aliph. CH_2 in six-ring in *rac*-**4**), 28.66 (2 aliph. CH_2 in six-ring in *rac*-**4**), 27.47 (aliph. CH_2 in six-ring in **5**), 24.12 (aliph. CH_2 in six-ring in **5**) ppm. EIMS (70eV): calcd. $C_{13}H_{12}$ 168.0939; found 168.0944.

4,9-Dihydro-1*H*-fluorene (5): By applying the General Procedure 3, a mixture of 4,4a-dihydro-1*H*-fluorene (*rac*-**4**) and 4,9-dihydro-1*H*-fluorene (**5**) (0.058 g, 0.3 mmol) gave 0.0533 g (92%) of the title compound as a pale brown oil, which solidified when cooled down. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.42 (m, 1 H, arom. CH), 7.27 (m, 1 H, arom. CH), 7.22 (m, 1 H, arom. CH), 7.15 (m, 1 H, arom. CH), 5.95 (dm, J = 9.5 Hz, 1 H, olefin. CH in six-ring), 5.89 (dm, J = 9.5 Hz, 1 H, olefin. CH in six-ring), 3.29 (s, 2 H, aliph. CH_2 in five-ring), 3.13 (s, 4 H, 2 aliph. CH_2 in six-ring) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 145.59 (C_q), 142.77 (C_q), 138.05 (C_q), 133.53 (C_q), 126.25 (arom. CH), 124.59 (olefin. CH in six-ring), 124.29 (arom. CH), 124.13 (olefin. CH in six-ring), 123.47 (arom. CH), 117.90 (arom. CH), 40.42 (aliph. CH_2 in five-ring), 27.43 (aliph. CH_2 in six-ring), 24.13 (aliph. CH_2 in six-ring) ppm. EIMS (70eV): calcd. $C_{13}H_{12}$ 168.0939; found 168.0942.

A Mixture of 2-Allyl-1-but-3-enyl-1*H*-indene (*rac*-6) and 1-But-3-enyl-2-[(*E*)-propenyl]-1*H*-indene (*rac*-7): By applying the General Procedure 1, 2-allyl-1*H*-indene (0.7708 g, 4.9 mmol), *n*BuLi (2.0 mL, 5.0 mmol, 2.5 M in *n*-hexane) and 4-bromo-1-butene (530 μL, 5.2 mmol) gave, after a 1-h reaction time, 0.9259 g (89%) of a 1:0.15 mixture of the title compounds as a colorless oil. ^{1}H NMR (500 MHz, 25 °C, $CDCl_3$): δ = 7.44 (m, 2 H, 2 arom. CH in *rac*-**7**), 7.42 (d, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**6**), 7.33 (m, 2 H, 2 arom. CH in *rac*-**7**), 7.31 (d, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**6**), 7.28 (m, 2 H, 2 arom. CH in *rac*-**7**), 7.27 (t, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**6**), 7.20 (m, 2 H, 2 arom. CH in *rac*-**7**), 7.18 (t, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**6**), 6.64 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**7**), 6.57 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**6**), 6.44 (d, J = 16.2 Hz, 2 H, 2 olefin. CH in chain in *rac*-**7**), 6.02 [a X-part of an ABX – system, dddd, J = 17.1 Hz, 10.2 Hz, 7.2 Hz (J_{AAX})*, 6.5 Hz (J_{AAX})*, 2 H, 2 olefin. CH in chain in *rac*-**6**], 5.96 (m, 2 H, 2 olefin. CH in chain in *rac*-**7**), 5.81 (ddt, J =

17.1 Hz, 10.2 Hz, 7.2 Hz, 2 H, 2 olefin. CH in chain in *rac*-**6**), 5.78 (m, 2 H, 2 olefin. CH in chain in *rac*-**7**), 5.23 (dm, J = 17.1 Hz, 2 H, 2 olefin. CH in the chain in *rac*-**6**), 5.17 (dm, J = 10.2 Hz, 2 H, 2 olefin. CH in the chain in *rac*-**6**), 5.02 (dm, J = 17.1 Hz, 2 H, 2 olefin. CH in the chain in *rac*-**6**), 4.98 (m, 4 H, 2 olefin. CH_2 in chain in *rac*-**7**), 4.97 (dm, J = 10.2 Hz, 2 H, 2 olefin. CH in the chain in *rac*-**6**), 3.72 (t, J = 5.2 Hz, 2 H, 2 aliph. CH in five-ring in *rac*-**7**), 3.50 (t, J = 5.2 Hz, 2 H, 2 aliph. CH in five-ring in *rac*-**6**), 3.30–3.23 (A-part of an ABX – system, dd, J_{AB} = –16.3 Hz, J_{AAX} = 6.5 Hz, 2 H, 2 aliph. CH in chain in *rac*-**6**)*, 3.17–3.10 (a B-part of an ABX – system, dd, J_{AB} = –16.3 Hz, J_{AAX} = 7.2 Hz, 2 H, 2 aliph. CH in chain in *rac*-**6**)*, 2.16 (m, 4 H, 2 aliph. CH in chain in *rac*-**6** and 2 aliph. CH in chain in *rac*-**7**), 1.91 (m, 14 H, 2 CH_3 in *rac*-**7**, 4 aliph. CH in chain in *rac*-**6** and 4 aliph. CH in chain in *rac*-**7**), 1.75 (m, 4 H, 2 aliph. CH in chain in *rac*-**6** and 2 aliph. CH in chain in *rac*-**7**). ^{13}C NMR (125.8 MHz, 25 °C, $CDCl_3$): δ = 151.35 (2 C_q in *rac*-**6**), 149.58 (2 C_q in *rac*-**7**), 147.83 (2 C_q in *rac*-**7**), 147.09 (2 C_q in *rac*-**6**), 144.91 (2 C_q in *rac*-**6**), 144.68 (2 C_q in *rac*-**7**), 138.98 (2 olefin. CH in chain in *rac*-**7**), 138.85 (2 olefin. CH in chain in *rac*-**6**), 136.10 (2 olefin. CH in chain in *rac*-**6**), 127.98 (2 olefin. CH in five-ring in *rac*-**7**), 127.74 (2 arom. CH in *rac*-**7**), 127.06 (2 olefin. CH in five-ring in *rac*-**6**), 127.02 (2 olefin. CH in chain in *rac*-**7**), 126.81 (2 olefin. CH in chain in *rac*-**7**), 126.62 (2 arom. CH in *rac*-**6**), 124.52 (2 arom. CH in *rac*-**7**), 124.03 (2 arom. CH in *rac*-**6**), 123.01 (2 arom. CH in *rac*-**7**), 122.93 (2 arom. CH in *rac*-**6**), 120.77 (2 arom. CH in *rac*-**7**), 120.27 (2 arom. CH in *rac*-**6**), 116.47 (2 olefin. CH_2 in chain in *rac*-**6**), 114.61 (2 olefin. CH_2 in chain in *rac*-**6**), 114.40 (2 olefin. CH_2 in chain in *rac*-**7**), 50.45 (2 aliph. CH in five-ring in *rac*-**6**), 48.00 (2 aliph. CH in five-ring in *rac*-**7**), 34.19 (2 aliph. CH_2 in chain in *rac*-**6**), 31.14 (2 aliph. CH_2 in chain in *rac*-**7**), 29.04 (2 aliph. CH_2 in chain in *rac*-**6**), 28.77 (2 aliph. CH_2 in chain in *rac*-**6**), 28.58 (2 aliph. CH_2 in chain in *rac*-**7**), 19.07 (2 CH_3 in *rac*-**7**). EIMS (70eV): calcd. $C_{16}H_{18}$ 210.1409; found 210.1413.

4b,5,6,9-Tetrahydrobenzo[*a*]azulene (*rac*-8): By applying the General Procedure 2, a mixture of 2-allyl-1-but-3-enyl-1*H*-indene (*rac*-**6**) and 1-but-3-enyl-2-[(*E*)-propenyl]-1*H*-indene (*rac*-**7**) (0.2539 g, 1.2 mmol) and Grubbs 2nd generation catalyst (0.053 g, 0.1 mmol) gave, after a 2-h reaction time, 0.1052 g (48%) of the title compound as a colorless oil. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.30 (d, J = 7.3 Hz, 2 H, 2 arom. CH), 7.22 (d, J = 7.3 Hz, 2 H, 2 arom. CH), 7.18 (t, J = 7.3 Hz, 2 H, 2 arom. CH), 7.10 (td, J = 7.3 Hz, 1.2 Hz, 2 H, 2 arom. CH), 6.39 (s, 2 H, 2 olefin. CH in five-ring), 5.82 (m, 2 H, 2 olefin. CH in seven-ring), 5.79 (m, 2 H, 2 olefin. CH in seven-ring), 3.37 (dd, J = 10.2 Hz, 5.7 Hz, 2 H, 2 aliph. CH in five-ring), 3.23 (m, 4 H, 2 aliph. CH_2 in seven-ring), 2.39 (dddd, J = –13.0 Hz, 7.4 Hz, 5.7 Hz, 2.1 Hz, 2 H, 2 aliph. CH in seven-ring), 2.22 (m, 2 H, 2 aliph. CH in seven-ring), 2.16 (m, 2 H, 2 aliph. CH in seven-ring), 1.23 (ddt, J = –13.0 Hz, 10.2 Hz, 2.0 Hz, 2 H, 2 aliph. CH in seven-ring). ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 149.98 (2 C_q), 148.12 (2 C_q), 144.51 (2 C_q), 132.14 (2 olefin. CH in seven-ring), 127.79 (2 olefin. CH in seven-ring), 126.64 (2 arom. CH), 125.52 (2 olefin. CH in five-ring), 123.98 (2 arom. CH), 122.38 (2 arom. CH), 120.22 (2 arom. CH), 54.59 (2 aliph. CH in five-ring), 30.92 (2 aliph. CH_2 in seven-ring), 30.33 (2 aliph. CH_2 in seven-ring), 25.83 (2 aliph. CH_2 in seven-ring). EIMS (70eV): calcd. $C_{14}H_{14}$ 182.1096; found 182.1099.

5,6,9,10-Tetrahydrobenzo[*a*]azulene (9): By applying the General Procedure 3, 4b,5,6,9-tetrahydro-benzo[*a*]azulene (*rac*-**8**) (0.0355 g, 0.2 mmol) gave 0.0343 g (97%) of the title compound as a yellow oil. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.34 (d, J = 7.3 Hz, 1 H, arom. CH), 7.24 (t, J = 7.3 Hz, 1 H, arom. CH), 7.18 (d, J = 7.3 Hz, 1 H, arom. CH), 7.10 (t, J = 7.3 Hz, 1 H, arom. CH), 5.96 (dtm, J = 10.6 Hz, 6.6 Hz, 1 H, olefin. CH in seven-ring), 5.84 (dt, J = 10.6 Hz, 5.2 Hz, 1 H, olefin. CH in seven-ring), 3.27 (m, 4 H, aliph. CH_2 in five-ring and aliph. CH_2 in seven-ring), 2.63 (m, 2 H, aliph. CH_2 in seven-ring), 2.45 (td, J = 6.4 Hz, 5.7 Hz, 2 H, aliph. CH_2 in seven-ring). ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 147.86 (C_q), 141.93 (C_q), 139.48 (C_q), 137.81 (C_q), 131.70 (olefin. CH in seven-ring), 128.98 (olefin. CH in seven-ring), 126.18 (arom. CH), 123.92 (arom. CH), 123.16 (arom. CH), 117.65 (arom. CH), 43.24 (aliph. CH_2 in five-ring), 29.72 (aliph. CH_2 in seven-ring), 26.02 (aliph. CH_2 in seven-ring), 24.55 (aliph. CH_2 in seven-ring). EIMS (70eV): calcd. $C_{14}H_{14}$ 182.1096; found 182.1101.

A Mixture of Allyl(2-allyl-1*H*-inden-1-yl)dimethylsilane (*rac*-10) and Allyldimethyl[(*E*)-2-propenyl]-1*H*-inden-1-yl]silane (*rac*-11): By applying the General Procedure 1, 2-allyl-1*H*-indene (0.3514 g, 2.3 mmol), *n*BuLi (910 μL, 2.3 mmol, 2.5 M in *n*-hexane) and allyl(chloro)dimethylsilane (350 μL, 2.4 mmol) gave, after a 1-h reaction time, 0.5177 g (90%) of a 1:0.13 mixture of the title compounds as a colorless oil. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.38 (d, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**10**), 7.37 (m, 4 H, 4 arom. CH in *rac*-**11**), 7.36 (d, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**10**), 7.22 (m, 2 H, 2 arom. CH in *rac*-**11**), 7.22 (t, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**10**), 7.11 (m, 2 H, 2 arom. CH in *rac*-**11**), 7.11 (t, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**10**), 6.73 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**11**), 6.63 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**10**), 6.43 (d, J = 16.1 Hz, 2 H, 2 olefin. CH in chain in *rac*-**11**), 5.99 [a X-part of an ABX – system, ddt, J = 17.5 Hz, 10.3 Hz, 6.8 Hz ($J_{AAX,AAX}$), 2 H, 2 olefin. CH in chain in *rac*-**10**][1], 5.89 (dq, J = 16.1 Hz, 6.7 Hz, 2 H, 2 olefin. CH in chain in *rac*-**11**), 5.70 (m, 2 H, 2 olefin. CH in chain in *rac*-**11**), 5.70 (tt, J = 12.2 Hz, 8.4 Hz, 2 H, 2 olefin. CH in chain in *rac*-**10**), 5.16 (d, J = 17.5 Hz, 2 olefin. CH in the chain in *rac*-**10**), 5.13 (d, J = 10.3 Hz, 2 olefin. CH in the chain in *rac*-**10**), 4.87 (m, 4 H, 2 olefin. CH_2 in chain in *rac*-**11**), 4.87 (d, J = 12.2 Hz, 4 H, 2 olefin. CH_2 in chain in *rac*-**10**), 3.79 (s, 2 H, 2 aliph. CH in five-ring in *rac*-**11**), 3.53 (s, 2 H, 2 aliph. CH in five-ring in *rac*-**10**), 3.40–3.34 (A-part of an ABX – system, dd, J_{AB} = –16.4 Hz, J_{AAX} = 6.8 Hz, 2 H, 2 aliph. CH in chain in *rac*-**10**)*, 3.27–3.22 (a B-part of an ABX – system, dd, J_{AB} = –16.4 Hz, J_{AAX} = 6.8 Hz, 2 H, 2 aliph. CH in chain in *rac*-**10**)*, 1.89 (d, J = 6.7 Hz, 6 H, 2 C–CH_3 in *rac*-**11**), 1.51 (m, 8 H, 2 aliph. CH_2 in chain in *rac*-**10** and 2 aliph. CH_2 in chain in *rac*-**11**), 0.05 (s, 6 H, Si–CH_3 in *rac*-**10**), –0.0 (s, 6 H, Si–CH_3 in *rac*-**10**), –0.03 (s, 6 H, Si–CH_3 in *rac*-**11**), –0.12 (s, 6 H, Si–CH_3 in *rac*-**11**). ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 150.16 (2 C_q in *rac*-**10**), 148.85 (2 C_q in *rac*-**11**), 145.07 (2 C_q in *rac*-**10**), 144.75 (2 C_q in *rac*-**10**), 144.45 (2 C_q in *rac*-**11**), 2 C_q in *rac*-**11** are overlapping with one of these signals, 136.50 (2 olefin. CH in chain in *rac*-**10** and 2 olefin. CH in chain *rac*-**11**), 134.70 (2 olefin. CH in chain in *rac*-**11**), 134.36 (2 olefin. CH in chain in *rac*-**10**), 127.95 (2 olefin. CH in chain in *rac*-**11** or 2 olefin. CH in five-ring in *rac*-**11**), 127.69 (2 olefin. CH in chain in *rac*-**11** or 2 olefin. CH in five-ring in *rac*-**11**), 125.77 (2 olefin. CH in five-ring *rac*-**10**), 125.20 (2 arom. CH in *rac*-**11**), 125.11 (2 arom. CH in *rac*-**10**), 123.34 (2 arom. CH in *rac*-**11**), 123.16 (2 arom. CH in *rac*-**10**), 123.03 (2 arom. CH in *rac*-**11**), 122.88 (2 arom. CH in *rac*-**10**), 120.77 (2 arom. CH in *rac*-**11**), 120.33 (2 arom. CH in *rac*-**10**), 116.29 (2 olefin. CH_2 in chain in *rac*-**10**), 114.00 (2 olefin. CH_2 in chain in *rac*-**10**), 133.81 (2 olefin. CH_2 in chain in *rac*-**11**), 46.71 (2 aliph. CH in five-ring in *rac*-**10**), 44.20 (2 aliph. CH in five-ring in *rac*-**11**), 36.24 (2 aliph. CH_2 in chain in *rac*-**10**), 22.01 (2 aliph. CH_2 in chain in *rac*-**10**), 21.89 (2 aliph. CH_2 in chain in *rac*-**11**), 18.69 (2 C–CH_3 in *rac*-**11**), –3.79 (2 Si–CH_3 in *rac*-**10**), –3.96 (2 Si–CH_3 in *rac*-**11**), –4.09 (2 Si–CH_3 in *rac*-**10**), –4.34 (2 Si–CH_3 in *rac*-**11**) ppm. ^{29}Si NMR (119.3 MHz,

25 °C, $CDCl_3$): δ = 3.36 (2 Si in *rac*-**11**), 3.02 (2 Si in *rac*-**10**) ppm. EIMS (70eV): calcd. $C_{17}H_{22}Si$ 254.1491; found 254.1498.

5,5-Dimethyl-4b,5,6,9-tetrahydro-5-silabenzo[*a*]azulene (*rac*-12): By applying the General Procedure 2, a mixture of allyl(2-allyl-1*H*-inden-1-yl)dimethylsilane (*rac*-**10**) and allyl(dimethyl){[(*E*)-2-propenyl]-1*H*-inden-1-yl}silane (*rac*-**11**) (0.2118 g, 0.8 mmol) and Grubbs 2nd generation catalyst (0.0359 g, 0.04 mmol) gave, after a 5-h reaction time, 0.1309 g (70%) of the title compound as a colorless oil. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.39 (d, J = 7.7 Hz, 4 H, 4 arom.), 7.22 (t, J = 7.7 Hz, 2 H, 2 arom. CH), 7.13 (t, J = 7.7 Hz, 2 H, 2 arom. CH), 6.55 (s, 2 H, 2 olefin. CH in five-ring), 5.78 (m, 4 H, 4 olefin. CH in seven-ring), 3.51 (s, 2 H, 2 aliph. CH in five-ring), 3.34–3.30 (A-part of an ABX – system, dd, J_{AB} = –14.7 Hz, J_{AAX} = 7.5 Hz, 2 H, 2 aliph. CH in seven-ring)*, 3.28–3.24 (a B-part of an ABX – system, dd, J_{AB} = –14.7 Hz, J_{AAX} = 5.2 Hz, 2 H, 2 aliph. CH in seven-ring)*, 1.94–1.91 (A-part of an ABX – system, dd, J_{AB} = –14.0 Hz, J_{AAX} = 5.3 Hz, 2 H, 2 aliph. CH in seven-ring)*, 1.53–1.50 (a B-part of an ABX – system, dd, J_{AB} = –14.0 Hz, J_{AAX} = 7.7 Hz, 2 H, 2 aliph. CH in seven-ring)*, 0.47 (s, 6 H, 2 CH_3), –0.74 (s, 6 H, 2 CH_3) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 146.84 (2 C_q), 145.13 (2 C_q), 144.92 (2 C_q), 128.03 (2 olefin. CH in seven-ring), 125.71 (2 olefin. CH in seven-ring), 124.86 (2 arom. CH), 123.77 (2 olefin. CH in five-ring), 122.81 (2 arom. CH), 121.75 (2 arom. CH), 120.18 (2 arom. CH), 50.37 (2 aliph. CH in five-ring), 29.07 (2 aliph. CH_2 in seven-ring), 16.72 (2 aliph. CH_2 in seven-ring), –1.30 (2 CH_3), –8.57 (2 CH_3) ppm. ^{29}Si NMR (119.3 MHz, 25 °C, $CDCl_3$): δ = 3.83 (2 Si) ppm. * Denotes: The spin-system is an ABX-system and the values of chemical shifts and coupling constants are approximations. EIMS (70eV): calcd. $C_{15}H_{18}Si$ 226.1178; found 226.1185.

A Mixture of a Diastereomeric Mixture of 1-Allyl-2-(1-methylallyl)-1*H*-indene (*rac*-13a and *rac*-13b) and 1-Allyl-2-[(*E*/*Z*)-propenyl]-1*H*-indene (*rac*-14): By applying the General Procedure 1, racemic 2-(1-methylallyl)-1*H*-indene (0.6555 g, 3.9 mmol), *n*BuLi (1.7 mL, 4.2 mmol, 2.5 M in *n*-hexane) and allyl bromide (510 µL, 5.8 mmol) gave, after a 2-h reaction time, 0.4463 g (55%) of a mixture of the title compounds as a colorless oil. The ratio between the diastereomeric mixture and the racemic mixture was 1:0.06. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.39 (d, J = 7.6 Hz, 2 H, 2 arom. CH in *rac*-**13a** or *rac*-**13b**), 7.38 (m, 2 H, 2 arom. CH in *rac*-**14**), 7.36 (d, J = 7.6 Hz, 2 H, 2 arom. CH in *rac*-**13a** or *rac*-**13b**), 7.23 (m, 12 H, 4 arom. CH in *rac*-**13a**, 4 arom. CH in *rac*-**13b**, 4 arom. CH in *rac*-**14**), 7.11 (t, J = 7.6 Hz, 4 H, 2 arom. CH in *rac*-**13a** and 2 arom. CH in *rac*-**13b**), 7.11 (m, 2 H, 2 arom. CH in *rac*-**14**), 6.65 (s, 2 H, 2 olefin. CH in *rac*-**14**), 6.54 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**13a** or *rac*-**13b**), 6.46 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**13a** or *rac*-**13b**), 6.05 (ddd, J = 16.8 Hz, 10.1 Hz, 7.2 Hz, 2 H, 2 olefin. CH in chain in *rac*-**13a** or *rac*-**13b**), 5.90 (ddm, J = 16.8 Hz, 10.1 Hz, 2 H, 2 olefin. CH in chain in *rac*-**14**), 5.79 (ddd, J = 16.8 Hz, 10.1 Hz, 7.2 Hz, 2 H, 2 olefin. CH in chain *rac*-**13a** or *rac*-**13b**), 5.43 (m, 4 H, 2 olefin. CH in *rac*-**13a** and 2 olefin. CH in *rac*-**13b**), 5.01 (m, 22 H, 4 olefin. CH_2 in chain in *rac*-**13a**, 4 olefin. CH_2 in chain in *rac*-**13b**, 2 olefin. CH in chain in *rac*-**14** and 2 olefin. CH_2 in chain in *rac*-**14**), 3.71 (dd, J = 7.2 Hz, 3.6 Hz, 2 H, 2 aliph. CH in five-ring in *rac*-**14**), 3.51 (t, J = 5.5 Hz, 2 H, 2 aliph. CH in five-ring *rac*-**13a** or *rac*-**13b**), 3.47 (t, J = 5.5 Hz, 2 H, 2 aliph. CH in five-ring *rac*-**13a** or *rac*-**13b**), 3.22 (m, 4 H, 2 aliph. CH in chain in *rac*-**13a** and 2 aliph. CH in chain in *rac*-**13b**), 2.79 (m, 6 H, 2 aliph. CH in chain in *rac*-**13a**, 2 aliph. CH in chain in *rac*-**13b** and 2 aliph. CH in chain in *rac*-**14**), 2.47 (dtm, J = –13.5 Hz, 5.5 Hz, 4 H, 2 aliph. CH in chain in *rac*-**13a** and 2 aliph. CH in chain in *rac*-**13b**), 2.34 (dtm, J = –14.3 Hz, 7.2 Hz, 2 H, 2 aliph. CH in chain in *rac*-**14**), 1.95 (s, 6 H, 2 CH_3 in chain in *rac*-**14**), 1.84 (d, J = 6.9 Hz, 6 H, 2 CH_3 in the end of the chain *rac*-**14**), 1.35 (d, J = 6.9 Hz, 6 H, 2 CH_3 in chain in *rac*-**13a** or *rac*-**13b**), 1.27 (d, J = 6.9 Hz, 6 H, 2 CH_3 in chain in *rac*-**13a** or *rac*-**13b**) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = (Cq in *rac*-**13a/b** and *rac*-**14** are partly overlapping with each other and other signals) 156.74 (C_q in *rac*-**13a/b**), 155.93 (C_q in *rac*-**13a/b**), 153.75 (C_q in *rac*-**14**), 153.11 (C_q in *rac*-**14**), 147.53 (C_q in *rac*-**14**), 147.10 (C_q in *rac*-**13a/b**), 146.71 (C_q in *rac*-**13a/b**), 145.50 (C_q in *rac*-**14**), 144.55 (C_q in *rac*-**13a/b**),143.42 (2 olefin. CH in chain in *rac*-**13a** or *rac*-**13b**), 143.31 (C_q in *rac*-**14**), 142.69 (2 olefin. CH in chain in *rac*-**14**), 141.69 (2 olefin. CH in chain in *rac*-**13a** or *rac*-**13b**), 135.04 (2 olefin. CH in chain in *rac*-**14**), 134.93 (2 olefin. CH in chain in *rac*-**13a** and 2 olefin. CH in chain in *rac*-**13b**), 126.72 (2 arom. CH in *rac*-**13a** or *rac*-**13b**), 126.65 (2 arom. CH in *rac*-**13a** or *rac*-**13b**), 126.41 (2 arom. CH in *rac*-**14**), 125.81 (2 olefin. CH in five-ring in *rac*-**13a** or *rac*-**13b**), 125.41 (2 olefin. CH in five-ring in *rac*-**14**), 125.32 (2 olefin. CH in five-ring in *rac*-**13a** or *rac*-**13b**), 124.25 (2 arom. CH in *rac*-**14**), 124.06 (2 arom. CH in *rac*-**13a** or *rac*-**13b**), 123.99 (2 arom. CH in *rac*-**13a** or *rac*-**13b**), 123.34 (2 arom. CH in *rac*-**14**), 123.21 (2 arom. CH in *rac*-**13a** and 2 arom. CH in *rac*-**13b**), 120.72 (2 arom. CH in *rac*-**14**), 120.40 (2 arom. CH in *rac*-**13a** and 2 arom. CH in *rac*-**13b**), 116.79 (2 olefin. CH_2 in chain in *rac*-**13a** or *rac*-**13b**), 116.64 (2 olefin. CH_2 in chain in *rac*-**13a** or *rac*-**13b**), 116.43 (2 olefin. CH_2 in chain in *rac*-**14**), 113.84 (2 olefin. CH_2 in chain in *rac*-**13a** or *rac*-**13b**), 113.61 (2 olefin. CH_2 in chain in *rac*-**13a** or *rac*-**13b**), 49.56 (2 aliph. CH in five-ring in *rac*-**13a** or *rac*-**13b**), 49.46 (2 aliph. CH in five-ring in *rac*-**13a** or *rac*-**13b**), 47.86 (2 aliph. CH in five-ring in *rac*-**14**), 38.08 (2 aliph. CH in chain in *rac*-**13a** or *rac*-**13b**), 37.53 (2 aliph. CH in chain in *rac*-**13a** or *rac*-**13b**), 37.10 (2 aliph. CH_2 in chain in *rac*-**14**), 34.48 (2 aliph. CH_2 in chain in *rac*-**13a** or *rac*-**13b**), 34.30 (2 aliph. CH_2 in chain in *rac*-**13a** or *rac*-**13b**), 20.82 (2 CH_3 in chain in *rac*-**13a** or *rac*-**13b**), 18.94 (2 CH_3 in chain in *rac*-**13a** or *rac*-**13b**), 14.52 (2 CH_3 in chain in *rac*-**14**), 14.42 (2 CH_3 in chain in *rac*-**14**) ppm. EIMS (70eV): calcd. $C_{16}H_{18}$ 210.1409; found 210.1400.

Diastereomeric Mixture of 1-Methyl-4,4a-dihydro-1*H*-fluorene (*rac*-15a and *rac*-15b): By applying the General Procedure 2, a mixture of 1-allyl-2-(1-methylallyl)-1*H*-indene (*rac*-**13a and** *rac*-**13b**) and 1-allyl-2-[(*E*/*Z*)-propenyl]-1*H*-indene (*rac*-**14**) (0.1837 g, 0.9 mmol) and Grubbs 2nd generation catalyst (0.0376 g, 0.04 mmol) gave, after a 4-h reaction time, 0.1091 g (69%) of a mixture of the title compounds as a colorless oil. The ratio between the diastereomers *rac*-**15a** and *rac*-**15b** was 1:0.9. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.38 (d, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**15a** or *rac*-**15b**), 7.36 (d, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**15a** or *rac*-**15b**), 7.31 (d, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**15a** or *rac*-**15b**), 7.30 (d, J = 7.3 Hz, 2 H, 2 arom. CH in *rac*-**15a** or *rac*-**15b**), 7.22 (t, J = 7.3 Hz, 4 H, 2 arom. CH in *rac*-**15a** and 2 arom. CH in *rac*-**15b**), 7.11 (t, J = 7.3 Hz, 4 H, 2 arom. CH in *rac*-**15a** and 2 arom. CH in *rac*-**15b**), 6.50 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**15a**), 6.46 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**15b**), 5.71 (m, 8 H, 8 olefin. CH in six-ring in *rac*-**15a** and *rac*-**15b**), 3.30 (m, 8 H, 4 aliph. CH in five-ring in *rac*-**15a** and *rac*-**15b** and 4 aliph. CH in six-ring in *rac*-**15a** and *rac*-**15b**), 2.85 (m, 4 H, 4 aliph. CH in six-ring in *rac*-**15a** and *rac*-**15b**), 1.71 (m, 4 H, 4 aliph. CH in six-ring in *rac*-**15a** and *rac*-**15b**), 1.32 (d, J = 7.1 Hz, 6 H, 2 CH_3 in *rac*-**15a** or *rac*-**15b**), 1.24 (d, J = 7.1 Hz, 6 H, 2 CH_3 in *rac*-**15a** or *rac*-**15b**). ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 155.04 (2 C_q in *rac*-**15a** or *rac*-**15b**), 154.95 (2 C_q in *rac*-**15a** or *rac*-**15b**), 147.80 (2 C_q in *rac*-**15a** or *rac*-**15b**), 147.57 (2 C_q in *rac*-**15a** or *rac*-**15b**), 144.94 (2 C_q in *rac*-**15a** or *rac*-**15b**), 144.85 (2 C_q in *rac*-**15a** or *rac*-**15b**), 132.57 (2 arom. CH in *rac*-**15a** or *rac*-**15b**), 132.48 (2 arom. CH in *rac*-**15a** or *rac*-**15b**), 126.72 (2 arom. CH in *rac*-**15a** or *rac*-**15b**), 126.68 (2

arom. CH in *rac*-**15a** or *rac*-**15b**), 124.91 (2 olefin. CH in six-ring in *rac*-**15a** or *rac*-**15b**), 124.64 (2 olefin. CH in six-ring in *rac*-**15a** or *rac*-**15b**), 123.87 (2 arom. CH in *rac*-**15a** or *rac*-**15b**), 123.81 (2 arom. CH in *rac*-**15a** or *rac*-**15b**), 122.96 (2 olefin. CH in six-ring in *rac*-**15a** or *rac*-**15b**), 122.83 (2 olefin. CH in six-ring in *rac*-**15a** or *rac*-**15b**), 122.49 (2 olefin. CH in five-ring in *rac*-**15a** or *rac*-**15b**), 121.69 (2 olefin. CH in five-ring in *rac*-**15a** or *rac*-**15b**), 120.57 (2 arom. CH in *rac*-**15a** and 2 arom. CH in *rac*-**15b**), 46.74 (2 aliph. CH in five-ring in *rac*-**15a** or *rac*-**15b**), 43.40 (2 aliph. CH in five-ring in *rac*-**15a** or *rac*-**15b**), 33.63 (2 aliph. CH in six-ring in *rac*-**15a** or *rac*-**15b**), 33.00 (2 aliph. CH in six-ring in *rac*-**15a** or *rac*-**15b**), 30.89 (2 aliph. CH in six-ring in *rac*-**15a** or *rac*-**15b**), 30.83 (2 aliph. CH in six-ring in *rac*-**15a** or *rac*-**15b**), 24.28 (2 CH_3 in *rac*-**15a** or *rac*-**15b**), 18.52 (2 CH_3 in *rac*-**15a** or *rac*-**15b**). EIMS (70eV): calcd. $C_{14}H_{14}$ 182.1096; found 182.1093.

1-Methyl-4,9-dihydro-1*H*-fluorene (*rac*-16): By applying the General Procedure 3, the diastereomeric mixture of 1-methyl-4,4a-dihydro-1*H*-fluorene (*rac*-**15a** and *rac*-**15b**) (0.043 g, 0.2 mmol) gave 0.0415 g (97%) of a mixture of the title compounds as a yellow oil. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.50 (d, J = 7.3 Hz, 2 H, 2 arom. CH), 7.35 (t, J = 7.3 Hz, 2 H, 2 arom. CH), 7.29 (d, J = 7.3 Hz, 2 H, 2 arom. CH), 7.23 (t, J = 7.3 Hz, 2 H, 2 arom. CH), 5.97 (m, 2 H, 2 olefin. CH in six-ring), 5.85 (m, 2 H, 2 olefin. CH in six-ring), 3.52 (d, J_{AB} = –21.9 Hz, 2 H, 2 aliph. CH in five-ring), 3.31 (m, 2 H, 2 aliph. CH in six-ring), 3.29 (d, J_{AB} = –21.9 Hz, 2 H, 2 aliph. CH in five-ring), 3.17 (m, 4 H, 2 aliph. CH_2 in six-ring), 1.34 (d, J = 7.1 Hz, 6 H, 2 CH_3). ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 145.51 (2 C_q), 143.22 (2 C_q), 142.74 (2 C_q), 132.91 (2 C_q), 131.38 (2 olefin. CH in six-ring), 126.31 (2 arom. CH), 124.15 (2 arom. CH), 123.56 (2 arom. CH), 123.02 (2 olefin. CH in six-ring), 118.14 (2 arom. CH), 38.56 (2 aliph. CH_2 in five-ring), 31.98 (2 aliph. CH in six-ring), 24.29 (2 aliph. CH_2 in six-ring), 21.89 (2 CH_3). EIMS (70eV): calcd. $C_{14}H_{14}$ 182.1096; found 182.1096.

Diastereomeric Mixture of 1-Allyl-2-(1-phenylallyl)-1*H*-indene (*rac*-17a and *rac*-17b): By applying the General Procedure 1, racemic 2-(1-phenylallyl)-1*H*-indene (0.6499 g, 2.8 mmol), *n*BuLi (1.3 mL, 3.1 mmol, 2.5 M in *n*-hexane) and allyl bromide (370 μL, 4.2 mmol) gave, after a 2-h reaction time, 0.4867 g (64%) of a mixture of the title compounds as a yellow oil. The ratio between the diastereomers *rac*-**17a** and *rac*-**17b** was 1:0.7. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.29 (m, 36 H, arom. CH in *rac*-**17a** and *rac*-**17b**), 6.76 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**17a**), 6.30 (ddd, J = 17.6 Hz, 10.1 Hz, 7.2 Hz, 2 H, 2 olefin. CH in chain in *rac*-**17a**), 6.22 (ddd, J = 17.6 Hz, 10.1 Hz, 8.0 Hz, 2 H, 2 olefin. CH in chain in *rac*-**17b**), 6.15 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**17b**), 5.41 (m, 4 H, 2 olefin. CH in chain in *rac*-**17a** and 2 olefin. CH in chain in *rac*-**17b**), 5.09 (m, 16 H, 2 olefin. CH_2 in chain in *rac*-**17a** and 2 olefin. CH_2 in chain in *rac*-**17b**), 4.48 (d, J = 8.0 Hz, 2 H, 2 aliph. CH in chain in *rac*-**17b**), 4.43 (d, J = 7.2 Hz, 2 H, 2 aliph. CH in chain in *rac*-**17a**), 3.64 (t, J = 5.2 Hz, 2 H, 2 aliph. CH in five-ring in *rac*-**17a**), 3.28 (t, J = 5.2 Hz, 2 H, 2 aliph. CH in five-ring in *rac*-**17b**), 2.88 (ddm, J = –14.5 Hz, 5.2 Hz, 2 H, 2 aliph. CH in chain in *rac*-**17b**), 2.75 (ddm, J = –14.5 Hz, 5.2 Hz, 2 H, 2 aliph. CH in chain in *rac*-**17a**), 2.57 (ddm, J = –14.5 Hz, 5.2 Hz, 2 H, 2 aliph. CH in chain in *rac*-**17b**), 2.50 (ddm, J = –14.5 Hz, 5.2 Hz, 2 H, 2 aliph. CH in chain in *rac*-**17a**) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 154.58 (2 C_q), 154.18 (2 C_q), 147.03 (4 C_q), 144.30 (4 C_q), 142.61 (2 C_q), 142.06 (2 C_q), 140.32 (2 olefin. CH in chain in *rac*-**17a**), 140.09 (2 olefin. CH in chain in *rac*-**17b**), 134.93 (2 olefin. CH in chain in *rac*-**17b**), 134.72 (2 olefin. CH in chain in *rac*-**17a**), 129.19 (2 olefin. CH in five-ring in *rac*-**17b**), 128.78 (2 arom. CH in *rac*-**17a**), 128.66 (4 arom. CH in *rac*-**17a**), 128.56 (2 arom. CH in *rac*-**17a**), 127.87 (2 olefin. CH in five-ring in *rac*-**17a**), 126.76 (2 arom. CH in *rac*-**17b**), 124.33 (2 arom. CH in *rac*-**17b**), 123.25 (2 arom. CH in *rac*-**17b**), 120.67 (2 arom. CH in *rac*-**17b**), 116.85 (2 olefin. CH_2 in chain in *rac*-**17a** or *rac*-**17b**), 116.79 (2 olefin. CH_2 in chain in *rac*-**17a** or *rac*-**17b**), 116.20 (2 olefin. CH_2 in chain in *rac*-**17a** or *rac*-**17b**), 115.71 (2 olefin. CH_2 in chain in *rac*-**17a** or *rac*-**17b**), 50.32 (2 aliph. CH in chain in *rac*-**17a**), 50.12 (2 aliph. CH in chain in *rac*-**17b**), 49.74 (2 aliph. CH in five-ring in *rac*-**17b**), 49.25 (2 aliph. CH in five-ring in *rac*-**17a**), 34.40 (2 aliph. CH_2 in chain in *rac*-**17b**), 34.33 (2 aliph. CH_2 in chain in *rac*-**17a**) ppm. EIMS (70eV): calcd. $C_{21}H_{20}$ 272.1565; found 272.1562.

Diastereomeric Mixture of 1-Phenyl-4,4a-dihydro-1*H*-fluorene (*rac*-18a and *rac*-18b): By applying the General Procedure 2, 1-allyl-2-(1-phenylallyl)-1*H*-indene (*rac*-**17a** and *rac*-**17b**) (0.1860 g, 0.7 mmol) and Grubbs 2nd generation catalyst (0.032 g, 0.03 mmol) gave, after a 4-h reaction time, 0.1493 g (90%) of a mixture of the title compounds as a pale yellow oil. The ratio between the diastereomers *rac*-**18a** and *rac*-**18b** was 1:0.8. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.39 (m, 36 H, arom. CH in *rac*-**18a** and *rac*-**18b**), 6.78 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**18b**), 6.19 (m, 2 H, 2 olefin. CH in six-ring in *rac*-**18b**), 6.12 (m, 2 H, 2 olefin. CH in six-ring in *rac*-**18a**), 6.09 (m, 2 H, 2 olefin. CH in six-ring in *rac*-**18b**), 6.02 (s, 2 H, 2 olefin. CH in five-ring in *rac*-**18a**), 5.96 (m, 2 H, 2 olefin. CH in six-ring in *rac*-**18a**), 4.69 (m, 2 H, 2 aliph. CH in six-ring in *rac*-**18b**), 4.58 (m, 2 H, 2 aliph. CH in six-ring in *rac*-**18a**), 3.61 (dd, J = 8.2 Hz, 7.8 Hz, 2 H, 2 aliph. CH in five-ring in *rac*-**18a**), 3.50 (dd, J = 8.2 Hz, 7.8 Hz, 2 H, 2 aliph. CH in five-ring in *rac*-**18b**), 3.09 (m, 4 H, 2 aliph. CH in six-ring in *rac*-**18a** and 2 aliph. CH in six-ring in *rac*-**18b**), 1.99 (m, 4 H, 2 aliph. CH in six-ring in *rac*-**18a** and 2 aliph. CH in six-ring in *rac*-**18b**) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 153.85 (2 C_q in *rac*-**18a**), 152.22 (2 C_q in *rac*-**18a**), 147.66 (2 C_q in *rac*-**18b**), 147.52 (2 C_q in *rac*-**18a**), 144.62 (2 C_q in *rac*-**18b**), 144.47 (2 C_q in *rac*-**18b**), 143.96 (2 C_q in *rac*-**18b**), 142.88 (2 C_q in *rac*-**18a**), 130.09 (2 olefin. CH in six-ring in *rac*-**18a**), 128.99 (2 olefin. CH in six-ring in *rac*-**18b**), 128.86, 128.62, 128.57, 127.74, 126.81, 126.73, 126.42, 124.04, 122.89, 122.84, 120.81, 120.74 (36 overlapping arom. CH in *rac*-**18a** and *rac*-**18b**), 127.35 (2 olefin. CH in six-ring in *rac*-**18b**), 126.13 (2 olefin. CH in six-ring in *rac*-**18a**), 124.87 (2 olefin. CH in five-ring in *rac*-**18a**), 123.61 (2 olefin. CH in five-ring in *rac*-**18b**), 46.79 (2 aliph. CH in five-ring in *rac*-**18a**), 45.64 (2 aliph. CH in six-ring in *rac*-**18a**), 44.45 (2 aliph. CH in six-ring in *rac*-**18b**), 43.59 (2 aliph. CH in five-ring in *rac*-**18b**), 30.76 (2 aliph. CH_2 in six-ring in *rac*-**18b**), 30.63 (2 aliph. CH_2 in six-ring in *rac*-**18a**) ppm. EIMS (70eV): calcd. $C_{19}H_{16}$ 244.1252; found 244.1251.

1-Phenyl-4,9-dihydro-1*H*-fluorene (*rac*-19): By applying the General Procedure 3, the diastereomeric mixture of 1-phenyl-4,4a-dihydro-1*H*-fluorene (*rac*-**18a** and *rac*-**18b**) (0.0927 g, 0.4 mmol) gave 0.0776 g (84%) of a mixture of the title compounds as a very thick, white oil. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.23 (m, 18 H, 18 arom. CH), 5.99 (m, 2 H, 2 olefin. CH in six-ring), 5.86 (m, 2 H, 2 olefin. CH in six-ring), 4.33 (m, 2 H, 2 aliph. CH in six-ring), 3.22 (m, 4 H, 2 aliph. CH_2 in six-ring), 3.14 (d, J_{AB} = –22.0 Hz, 2 H, 2 aliph. CH in five-ring), 3.01 (d, J_{AB} = –22.0 Hz, 2 H, 2 aliph. CH in five-ring) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 145.18 (2 C_q), 144.52 (2 C_q), 143.03 (2 C_q), 141.11 (2 C_q), 133.67 (2 C_q), 129.22 (2 olefin. CH in six-ring), 128.66 (4 arom. CH), 128.30 (4 arom. CH), 126.63 (2 arom. CH), 126.28 (2 arom. CH), 124.45 (2 arom. CH), 123.61 (2 arom. CH), 123.40 (2 olefin. CH in six-ring), 118.39 (2 arom. CH), 44.31 (2 aliph. CH in six-ring), 39.05 (2 aliph. CH_2 in five-ring), 24.27 (2 aliph. CH_2 in six-ring) ppm. EIMS (70eV): calcd. $C_{19}H_{16}$ 244.1252; found 244.1247.

A Mixture of 1,3-Bis[allyl(dimethyl)silanyl]-1*H*-indene (*rac*-20) and 1,1-Bis[allyl(dimethyl)silanyl]-1*H*-indene (21): By applying the Ge-

neral Procedure 4, indene (500 μL, 4.3 mmol), *n*BuLi (2 × 1.8 mL, 2×4.3 mmol, 2.5 M in *n*-hexane) and allyl(chloro)dimethylsilane (2×700 μL, 2×4.6 mmol) gave 0.6357 g (48%) of a mixture of the title compounds as a pale yellow oil. The ratio between the obtained compounds was 1:0.04. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): 7.56 (d, *J* = 7,4 Hz, 2 H, 2 arom. CH in *rac*-**20**), 7.53 (d, *J* = 7.4 Hz, 1 H, arom. CH in **21**), 7.49 (d, *J* = 7.4 Hz, 1 H, arom. CH in **21**), 7.47 (d, *J* = 7.4 Hz, 2 H, 2 arom. CH in *rac*-**20**), 7.26 (t, *J* = 7.4 Hz, 2 H, 2 arom. CH in *rac*-**20**), 7.26 (m, 1 H, arom. CH in **21**), 7.18 (t, *J* = 7.4 Hz, 2 H, 2 arom. CH in *rac*-**20**), 7.18 (m, 1 H, arom. CH in **21**), 7.05 (d, *J* = 5.2 Hz, 1 H, olefin. CH in five-ring in **21**), 6.88 (d, *J* = 1.6 Hz, 2 H, 2 olefin. CH in five-ring in *rac*-**20**), 6.69 (d, *J* = 5.2 Hz, 1 H, olefin. CH in five-ring in **21**), 5.81 (ddt, *J* = 18.1 Hz, 10.1 Hz, 8.2 Hz, 2 H, 2 olefin. CH in chain *rac*-**20**), 5.70 (ddt, *J* = 18.1 Hz, 10.1 Hz, 8.2 Hz, 2 H, 2 olefin. CH in chain *rac*-**20**), 5.45 [a X-part of an ABX – system, ddt, *J* = 18.1 Hz, 10.1 Hz, 8.2 Hz ($J_{AAX,AAX}$), 2 H, 2 olefin. CH in chain **21**][1], 4.87 (m, 8 H, 4 olefin. CH_2 in chain in *rac*-**20**), 4.72 (d, *J* = 10.1 Hz, 2 H, olefin. CH_2 in chain in **21**), 4.69 (d, *J* = 18.1 Hz, 2 H, olefin. CH_2 in chain in **21**), 3.67 (d, *J* = 1.6 Hz, 2 H, 2 aliph. CH in five-ring in *rac*-**20**), 1.80 (d, *J* = 8.2 Hz, 4 H, 2 aliph. CH_2 in chain in *rac*-**20**), 1.50 (d, *J* = 8.2 Hz, 4 H, 2 aliph. CH_2 in chain in *rac*-**20**), 1.29–1.24 (A-part of an ABX – system, dd, J_{AB} = –13.5 Hz, J_{AAX} = 8.2 Hz, 2 H, 2 aliph. CH in chain in **21**)*, 1.19–1.14 (a B-part of an ABX – system, dd, J_{AB} = –13.5 Hz, J_{AAX} = 8.2 Hz, 2 H, 2 aliph. CH in chain in **21**)*, 0.324 (s, 6 H, 2 CH_3 in *rac*-**20**), 0.321 (s, 6 H, 2 CH_3 in *rac*-**20**), 0.04 (s, 6 H, 2 CH_3 in **21**), –0.01 (s, 6 H, 2 CH_3 in **20**), –0.05 (s, 6 H, 2 CH_3 in *rac*-**20**), –0.13 (s, 6 H, 2 CH_3 in *rac*-**20**) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 147.52 (2 C_q in *rac*-**20**), 146.19 (2 C_q in *rac*-**20**), 146.05 (2 olefin. CH in five-ring in *rac*-**20**), 140.27 (2 C_q in *rac*-**20**), 3 C_q in **21** could not be detected due to the very small amount of the compound **21**, 2 olefin. CH in five-ring in **21** are overlapping with other signals, 137.19 (olefin. CH in chain in **21**), 135.07 (2 olefin. CH in chain in *rac*-**20**), 134.77 (olefin. CH in chain in **21**), 134.41 (2 olefin. CH in chain in *rac*-**20**), 124.84 (2 arom. CH in *rac*-**20**), 124.42 (arom. CH in **21**), 123.63 (2 arom. CH in *rac*-**20**), 123.40 (arom. CH in **21**), 122.95 (2 arom. CH in *rac*-**20**), 122.33 (2 arom. CH in *rac*-**20**), 121.53 (arom. CH in **21**), one arom. CH in **21** is overlapping with other signals, 113.86 (2 olefin. CH_2 in chain in *rac*-**20**), 113.54 (olefin. CH_2 in chain in **21**), 133.43 (2 olefin. CH_2 in chain in *rac*-**20**), olefin. CH_2 in chain in **21** in overlapping with other signals, 47.57 (2 aliph. CH in five-ring in *rac*-**20**), 23.89 (2 aliph. CH_2 in chain in *rac*-**20**), 22.52 (2 aliph. CH_2 in chain in *rac*-**20** and aliph. CH_2 in chain in **21**), 22.14 (aliph. CH_2 in chain in **21**), –2.54 (2 CH_3 in **21**), –2.84 (2 CH_3 in *rac*-**20**), –2.95 (2 CH_3 in *rac*-**20**), –3.14 (2 CH_3 in **21**), –4.41 (2 CH_3 in *rac*-**20**), –4.81 (2 CH_3 in *rac*-**20**) ppm. ^{29}Si NMR (119.3 MHz, 25 °C, $CDCl_3$): δ = 3.38 (2 Si in *rac*-**20**), 0.45 (2 Si in **21**), –9.98 (2 Si in *rac*-**20**) ppm. EIMS (70eV): calcd. $C_{19}H_{28}Si_2$ 312.1730; found 312.1728.

Spiro Compound 22: By applying General Procedure 2, a mixture of 1,3-bis[allyl(dimethyl)silanyl]-1*H*-indene (*rac*-**20**) and 1,1-bis[allyl(dimethyl)silanyl]-1*H*-indene (**21**) (0.1497 g, 0.5 mmol) and Grubbs 2nd generation catalyst (0.0222 g, 0.03 mmol) gave, after a 24-h reaction time, 0.023 g (17%) of the title compound as a colorless oil. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.66 (d, *J* = 7.7 Hz, 1 H, arom. CH), 7.51 (d, *J* = 7.7 Hz, 1 H, arom. CH), 7.24 (t, *J* = 7.7 Hz, 1 H, arom. CH), 7.21 (t, *J* = 7.7 Hz, 1 H, arom. CH), 7.03 (d, *J* = 5.0 Hz, 1 H, olefin. CH in five-ring), 6.82 (d, *J* = 5.0 Hz, 1 H, olefin. CH in five-ring), 5.68 (a X-part of an ABX – system, m, 2 H, 2 olefin. CH in seven-ring)*, 1.92–1.87 (A-part of an ABX – system, dd, J_{AB} = –14.2 Hz, J_{AAX} = 5.8 Hz, 2 H, 2 aliph. CH in seven-ring)*, 1.83–1.79 (a B-part of an ABX – system, dd, J_{AB} = –14.2 Hz, J_{AAX} = 6.0 Hz, 2 H, 2 aliph. CH in seven-ring)*, –0.27 (s, 6 H, 2 CH_3), –0.30 (s, 6 H, 2 CH_3) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 146.88 (C_q), 144.47 (C_q), 137.34 (olefin. CH in five-ring), 128.15 (olefin. CH in five-ring), 124.76 (2 olefin. CH in seven-ring), 124.15 (arom. CH), 123.39 (arom. CH), 123.08 (arom. CH), 121.25 (arom. CH), 50.96 (C_q), 17.69 (2 aliph. CH_2 in seven-ring), –3.47 (2 CH_3), –3.81 (2 CH_3) ppm. ^{29}Si NMR (119.3 MHz, 25 °C, $CDCl_3$): δ = –5.27 ppm. EIMS (70eV): calcd. $C_{17}H_{24}Si_2$ 284.1417; found 284.1415.

A Mixture of 1,3-Bis[(hex-5-enyl)dimethylsilanyl]-1*H*-indene (*rac*-23) and 1,1-Bis[(hex-5-enyl)dimethylsilanyl]-1*H*-indene (24): By applying the General Procedure 4, indene (810 μL, 6.9 mmol), *n*BuLi (2 × 2.8 mL, 2×7.0 mmol, 2.5 M in *n*-hexane) and chloro(5-hexenyl)dimethylsilane (2 × 1.5 mL, 2×7.6 mmol) gave 2.1314 g (78%) of a mixture of the title compounds as a yellow oil. The ratio between the obtained compounds was 1:0.07. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.66 (d, *J* = 7.5 Hz, 1 H, arom. CH in **24**), 7.62 (d, *J* = 7.5 Hz, 2 H, 2 arom. CH in *rac*-**23**), 7.59 (d, *J* = 7.5 Hz, 1 H, arom. CH in **24**), 7.53 (d, *J* = 7.5 Hz, 2 H, 2 arom. CH in *rac*-**23**), 7.32 (t, *J* = 7.5 Hz, 2 H, 2 arom. CH in *rac*-**23**), 7.32 (m, 1 H, arom. CH in **24**), 7.24 (d, *J* = 7.5 Hz, 2 H, 2 arom. CH in *rac*-**23**), 7.24 (m, 1 H, arom. CH in **24**), 7.09 (d, *J* = 5.1 Hz, 1 H, olefin. CH in five-ring in **24**), 6.91 (m, 2 H, 2 olefin. CH in five-ring in *rac*-**23**), 6.76 (d, *J* = 5.1 Hz, 1 H, olefin. CH in five-ring in **24**), 5.85 (ddt, *J* = 16.9 Hz, 10.3 Hz, 6.8 Hz, 4 H, 4 olefin. CH in chain in *rac*-**23**), 5.47 (m, 6 H, 2 olefin. CH and 2 olefin. CH_2 in chain in **24**), 5.07 (dm, *J* = 10.3 Hz, 2 H, 2 olefin. CH in chain in *rac*-**23**), 5.03 (dm, *J* = 16.9 Hz, 2 H, 2 olefin. CH in chain in *rac*-**23**), 5.02 (dm, *J* = 16.9 Hz, 2 H, 2 olefin. CH in chain in *rac*-**23**), 4.99 (dm, *J* = 10.3 Hz, 2 H, 2 olefin. CH in chain in *rac*-**23**), 3.71 (m, 2 H, 2 aliph. CH in five-ring in *rac*-**23**), 2.11 (m, 8 H, 4 aliph. CH_2 in chain in *rac*-**23**), 2.02 (m, 8 H, 2 aliph. CH_2 in chain in **24**), 1.70 (m, 4 H, 2 aliph. CH_2 in chain in **24**), 1.41 (m, 8 H, 4 aliph. CH_2 in chain in *rac*-**23**), 0.97 (t, *J* = 6.6 Hz, 4 H, 2 aliph. CH_2 in chain in **24**), 0.91 (dd, *J* = 6.9 Hz, 6.6 Hz, 4 H, 2 aliph. CH_2 in chain in *rac*-**23**), 0.56 (t, *J* = 8.3 Hz, 4 H, 2 aliph. CH_2 in chain in *rac*-**23**), 0.38 (s, 12 H, 4 CH_3 in *rac*-**23**), 0.12 (s, 6 H, 2 CH_3 in **24**), 0.05 (s, 6 H, 2 CH_3 **24**), –0.00 (s, 6 H, 2 CH_3 in *rac*-**23**), –0.05 (s, 6 H, 2 CH_3 in *rac*-**23**) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = Intensity of the C_q – signals in **24** are so low that they could not be observed, 147.80 (2 C_q in *rac*-**23**), 146.48 (2 C_q in *rac*-**23**), 145.92 (2 olefin. CH in five-ring in *rac*-**23**), 140.64 (2 C_q in *rac*-**23**), 139.26 (2 olefin. CH in chain in **24**), 139.21 (2 olefin. CH in chain in *rac*-**23**), 139.05 (2 olefin. CH in chain in *rac*-**23**), 124.86 (arom. CH in **24**), 124.64 (2 arom. CH in *rac*-**23**), 123.99 (arom. CH in **24**), 123.39 (2 arom. CH in *rac*-**23**), 123.10 (arom. CH in **24**), 122.85 (2 arom. CH in *rac*-**23**), 122.22 (arom. CH in **24**), 122.15 (2 arom. CH in *rac*-**23**), 144.42 (2 aliph. CH_2 in chain in *rac*-**23**), 114.25 (2 aliph. CH_2 in chain in *rac*-**23**), 2 aliph. CH_2 in chain in **24** are overlapping with other signals, 47.94 (2 aliph. CH in five-ring in *rac*-**23**), 33.68 (2 aliph. CH_2 in chain in **24**), 33.64 (2 aliph. CH_2 in chain in *rac*-**23**), 33.56 (2 aliph. CH_2 in chain in *rac*-**23**), 32.94 (2 aliph. CH_2 in chain in *rac*-**23**), 32.81 (2 aliph. CH_2 in chain in *rac*-**23**), 32.72 (2 aliph. CH_2 in chain in **24**), 23.80 (2 aliph. CH_2 in chain in *rac*-**23**), 23.35 (2 aliph. CH_2 in chain in *rac*-**23**), 22.93 (2 aliph. CH_2 in chain in **24**), 18.38 (2 aliph. CH_2 in chain in **24**), 15.81 (2 aliph. CH_2 in chain in *rac*-**23**), 14.51 (2 aliph. CH_2 in chain in *rac*-**23**), 0.51 (2 CH_3 in **24**), –2.27 (2 CH_3 in **24** and 2 CH_3 in *rac*-**23**), –2.52 (2 CH_3 in *rac*-**23**), –4.00 (2 CH_3 in *rac*-**23**), –4.26 (2 CH_3 in *rac*-**23**) ppm. ^{29}Si NMR (119.3 MHz, 25 °C, $CDCl_3$): δ = 4.81 (2 Si in *rac*-**23**), 2.09 (Si in **24**), 1.65 (Si in **24**), –8.44 (2 Si in *rac*-**23**) ppm. EIMS (70eV): calcd. $C_{25}H_{40}Si_2$ 396.2669; found 396.2680.

A Mixture of (*E*)-2,2,12,12-Tetramethyl-2,12-disilatricyclo-[11.6.1.0*14,19*]icosa-1(20),7,14,16,18-pentaene [*rac*-(*E*)-25], (*Z*)-2,2,12,12-Tetramethyl-2,12-disilatricyclo[11.6.1.0*14,19*]icosa-1(20),7,14,16,18-pentaene [*rac*-(*Z*)-25] and (*Z*)-2,2,13,13-Tetramethyl-2,13-disilatricyclo[12.6.1.0*15,20*]henicosa-1(21),7,15,17,19-pentaene [*rac*-(*Z*)-26]: By applying the General Procedure 2, a mixture of 1,3-bis[(hex-5-enyl)dimethylsilanyl]-1*H*-indene (*rac*-**23**) and 1,1-bis[(hex-5-enyl)dimethylsilanyl]-1*H*-indene (**24**) (0.2422 g, 0.6 mmol) and Grubbs 2nd generation catalyst (0.0322 g, 0.04 mmol) gave, after a 22-h reaction time, 0.0226 g (10%) of a mixture of the title compounds as a colorless oil. The ratio between the obtained compounds was 0.8:0.16:0.15. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.54 [d, J = 7.5 Hz, 2 H, 2 arom. CH in *rac*-(*E*)-**25**], 7.54 [m, 4 H, 2 arom. CH in *rac*-(*Z*)-**25** and 2 arom. CH in *rac*-(*Z*)-**26**], 7.46 [d, J = 7.5 Hz, 2 H, 2 arom. CH in *rac*-(*E*)-**25**], 7.46 [m, 4 H, 2 arom. CH in *rac*-(*Z*)-**25** and 2 arom. CH in *rac*-(*Z*)-**26**], 7.25 [t, J = 7.5 Hz, 2 H, 2 arom. CH in *rac*-(*E*)-**25**], 7.25 [m, 4 H, 2 arom. CH in *rac*-(*Z*)-**25** and 2 arom. CH in *rac*-(*Z*)-**26**], 7.16 [t, J = 7.5 Hz, 2 H, 2 arom. CH in *rac*-(*E*)-**25**], 7.16 [m, 4 H, 2 arom. CH in *rac*-(*Z*)-**25** and 2 arom. CH in *rac*-(*Z*)-**26**], 6.91 [m, 2 H, 2 olefin. CH in five-ring in *rac*-(*Z*)-**26**], 6.84 [m, 4 H, 2 olefin. CH in five-ring in *rac*-(*E*)-**25** and 2 olefin. CH in five-ring in *rac*-(*Z*)-**25**], 5.39 [m, 4 H, 4 olefin. CH in large ring in *rac*-(*Z*)-**25**], 5.25 [m, 4 H, 4 olefin. CH in large ring in *rac*-(*Z*)-**26**], 4.98 [ddd, J = 14.3 Hz, 7.4 Hz, 7.0 Hz, 2 H, 2 olefin. CH in large ring in *rac*-(*E*)-**25**], 4.88 [ddd, J = 14.3 Hz, 7.4 Hz, 7.0 Hz, 2 H, 2 olefin. CH in large ring in *rac*-(*E*)-**25**], 3.63 [m, 2 H, 2 aliph. CH in five-ring in *rac*-(*Z*)-**25**], 3.60 [m, 2 H, 2 aliph. CH in five-ring in *rac*-(*E*)-**25**], 3.57 [m, 2 H, 2 aliph. CH in five-ring in *rac*-(*Z*)-**26**], 14 CH_2 in large ring in *rac*-(*Z*)-**25** and 8 CH_3 in *rac*-(*Z*)-**25** are overlapping with other signals, 2.08 [m, 4 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25** and 2 aliph. CH in large ring in *rac*-(*Z*)-**26**], 2.01 [m, 4 H, 4 aliph. CH in large ring in *rac*-(*Z*)-**26**], 1.92 [m, 10 H, 4 aliph. CH in large ring in *rac*-(*E*)-**25** and 6 aliph. CH in large ring in *rac*-(*Z*)-**26**], 1.63 [m, 2 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25**], 1.44 [m, 6 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25** and 4 aliph. CH in large ring in *rac*-(*Z*)-**26**], 1.32 [m, 6 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25** and 4 aliph. CH in large ring in *rac*-(*Z*)-**26**], 1.25 [m, 6 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25** and 4 aliph. CH in large ring in *rac*-(*Z*)-**26**], 1.16 [m, 2 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25**], 1.03 [m, 2 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25**], 0.88 [m, 2 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25**], 0.82 [m, 2 H, 2 aliph. CH in large ring in *rac*-(*Z*)-**26**], 0.72 [m, 2 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25**], 0.67 [m, 2 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25**], 0.48 [m, 2 H, 2 aliph. CH in large ring in *rac*-(*Z*)-**26**], 0.29 [m, 36 H, 6 CH_3 in *rac*-(*E*)-**25** and 6 CH_3 in *rac*-(*Z*)-**26**], 0.18 [s, 6 H, 2 CH_3 in *rac*-(*E*)-**25**], –0.11 [m, 6 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25** and 4 aliph. CH in large ring in *rac*-(*Z*)-**26**], –0.38 [s, 6 H, 2 CH_3 in *rac*-(*Z*)-**26**], –0.48 [m, 2 H, 2 aliph. CH in large ring in *rac*-(*E*)-**25**] ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 147.89 [2 C_q in *rac*-(*Z*)-**26**], 147.71 [2 C_q in *rac*-(*Z*)-**25**], 147.62 [2 C_q in *rac*-(*E*)-**25**], 146.71 [2 C_q in *rac*-(*E*)-**25**], 146.59 [2 olefin. CH in five-ring in *rac*-(*E*)-**25** and 2 olefin. CH in five-ring in *rac*-(*Z*)-**25** and 2 C_q in *rac*-(*Z*)-**26**], 146.29 [2 C_q in *rac*-(*Z*)-**25**], 146.09 [2 olefin. CH in five-ring in *rac*-(*Z*)-**26**], 140.53 [2 C_q in *rac*-(*Z*)-**25**], 140.32 [2 C_q in *rac*-(*E*)-**25**], 140.05 [2 C_q in *rac*-(*Z*)-**26**], 131.98 [2 olefin. CH in large ring in *rac*-(*E*)-**25**], 131.68 [2 olefin. CH in large ring in *rac*-(*Z*)-**25**], 131.53 [2 olefin. CH in large ring in *rac*-(*Z*)-**25**], 131.14 [2 olefin. CH in large ring in *rac*-(*Z*)-**26**], 130.98 [2 olefin. CH in large ring in *rac*-(*Z*)-**26**], 130.86 [2 olefin. CH in large ring in *rac*-(*E*)-**25**], 8 arom. CH in *rac*-(*Z*)-**25** are overlapping with other signals, 124.63 [2 arom. CH in *rac*-(*E*)-**25**], 124.60 [2 arom. CH in *rac*-(*Z*)-**26**], 123.30 [2 arom. CH in *rac*-(*E*)-**25**], 123.29 [2 arom. CH in *rac*-(*Z*)-**26**], 122.87 [2 arom. CH in *rac*-(*Z*)-**26**], 122.75 [2 arom. CH in *rac*-(*E*)-**25**], 122.16 [2 arom. CH in *rac*-(*E*)-**25** and 2 arom. CH in *rac*-(*Z*)-**26**], 49.07 [2 aliph. CH in five-ring in *rac*-(*Z*)-**25**], 48.79 [2 aliph. CH in five-ring in *rac*-(*Z*)-**26**], 48.52 [2 aliph. CH in five-ring in *rac*-(*E*)-**25**], 35.45 [2 aliph. CH_2 in large ring in *rac*-(*E*)-**25**], 35.12 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**25**], 33.55 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**26**], 32.74 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**26**], 32.54 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**26**], 32.37 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**25**], 32.25 [2 aliph. CH_2 in large ring in *rac*-(*E*)-**25**], 31.63 [2 aliph. CH_2 in large ring in *rac*-(*E*)-**25**], 31.22 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**26**], 30.60 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**25**], 22.72 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**25**], 22.47 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**26**], 22.44 [2 aliph. CH_2 in large ring in *rac*-(*E*)-**25**], 22.32 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**26**], 22.15 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**25**], 22.10 [2 aliph. CH_2 in large ring in *rac*-(*E*)-**25**], 15.90 [2 aliph. CH_2 in large ring in *rac*-(*E*)-**25**], 15.88 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**25**], 14.95 [2 aliph. CH_2 in large ring in *rac*-(*Z*)-**26**], 9.17 [2 aliph. CH_2 in large ring in *rac*-(*E*)-**25**, 2 aliph. CH_2 in large ring in *rac*-(*Z*)-**25** and 2 aliph. CH_2 in large ring in *rac*-(*Z*)-**26**], –1.19 [2 CH_3 in *rac*-(*Z*)-**26**], –1.32 [2 CH_3 in *rac*-(*E*)-**25** and 2 CH_3 in *rac*-(*Z*)-**25**], –1.72 [2 CH_3 in *rac*-(*E*)-**25** and 2 CH_3 in *rac*-(*Z*)-**25**], –1.84 [2 CH_3 in *rac*-(*Z*)-**26**], –1.92 [2 CH_3 in *rac*-(*Z*)-**26**], –3.45 [2 CH_3 in *rac*-(*E*)-**25** and 2 CH_3 in *rac*-(*Z*)-**25**], –3.47 [2 CH_3 in *rac*-(*E*)-**25** and 2 CH_3 in *rac*-(*Z*)-**25**], –5.71 [2 CH_3 in *rac*-(*Z*)-**26**] ppm. ^{29}Si NMR (119.3 MHz, 25 °C, $CDCl_3$): δ = 5.48 [2 Si in *rac*-(*Z*)-**25** or *rac*-(*Z*)-**26**], 4.92 [2 Si in *rac*-(*Z*)-**25** or *rac*-(*Z*)-**26**], 4.83 [2 Si in *rac*-(*E*)-**25**], –7.98 [2 Si in *rac*-(*E*)-**25**], –8.46 [2 Si in *rac*-(*Z*)-**25** or *rac*-(*Z*)-**26**], –8.63 [2 Si in *rac*-(*Z*)-**25** or *rac*-(*Z*)-**26**] ppm. EIMS (70eV): calcd. $C_{22}H_{34}Si_2$ 354.2199; found 354.2204 [*rac*-(*E*)-**25** and *rac*-(*Z*)-**25**], calcd. $C_{23}H_{36}Si_2$ 368.2356; found 368.2365 [*rac*-(*Z*)-**26**].

A Mixture of 1,3-Di(pent-4-enyl)-1*H*-indene (*rac*-27) and 1,1-Di-(pent-4-enyl)-1*H*-indene (28): By applying the General Procedure 4, indene (500 μL, 4.3 mmol), *n*BuLi (2 × 1.9 mL, 2×4.7 mmol, 2.5 M in *n*-hexane) and 5-bromo-1-pentene (2×560 μL, 2×4.7 mmol) gave 0.8044 g (75%) of a mixture of the title compounds as a colorless oil. The ratio between the obtained compounds was 1:0.23. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.41 (m, 2 H, 2 arom. CH in *rac*-**27**), 7.30 (m, 5 H, 4 arom. CH in *rac*-**27** and arom. CH in **28**), 7.21 (m, 5 H, 2 arom. CH in *rac*-**27** and 3 arom. CH in **28**), 6.72 (d, J = 6.0 Hz, 1 H, olefin. CH in five-ring in **28**), 6.29 (d, J = 6.0 Hz, 1 H, olefin. CH in five-ring in **28**), 6.23 (m, 2 olefin. CH in five-ring in *rac*-**27**), 5.89 (ddt, J = 16.8 Hz, 10.1 Hz, 6.7 Hz, 2 H, 2 olefin. CH in chain in *rac*-**27**), 5.82 (ddt, J = 16.8 Hz, 10.1 Hz, 6.7 Hz, 2 H, 2 olefin. CH in chain in *rac*-**27**), 5.69 (ddt, J = 16.8 Hz, 10.1 Hz, 6.7 Hz, 2 H, 2 olefin. CH in chain in **28**), 5.08 (dm, J = 16.8 Hz, 2 H, 2 olefin. CH in chain in *rac*-**27**), 5.03 (dm, J = 16.8 Hz, 2 H, 2 olefin. CH in chain in *rac*-**27**), 5.02 (dm, J = 10.1 Hz, 2 H, 2 olefin. CH in chain in *rac*-**27**), 4.97 (dm, J = 10.1 Hz, 2 H, 2 olefin. CH in chain in *rac*-**27**), 4.93 (dm, J = 16.8 Hz, 2 H, 2 olefin. CH in chain in **28**), 4.89 (dm, J = 16.8 Hz, 2 H, 2 olefin. CH in chain in **28**), 3.42 (m, 2 H, 2 aliph. CH in five-ring in *rac*-**27**), 2.56 (m, 4 H, 2 aliph. CH_2 in chain in *rac*-**27**), 2.15 (m, 8 H, 4 aliph. CH_2 in chain in *rac*-**27**), 1.92 (m, 4 H, 2 aliph. CH_2 in chain in *rac*-**27**), 1.77 (m, 8 H, 2 aliph. CH_2 in chain in *rac*-**27** and 2 aliph. CH_2 in chain in **28**), 1.51 (m, 8 H, 2 aliph. CH_2 in chain in *rac*-**27** and 2 aliph. CH_2 in chain in **28**), 1.19 (m, 2 H, 2 aliph. CH in chain in **28**), 0.93 (m, 2 H, 2 aliph. CH in chain in **28**) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 150.99 (C_q in **28**), 148.83 (2 C_q in *rac*-**27**), 145.17 (2 C_q in *rac*-**27**), 144.49 (C_q in **28**), 144.13 (olefin. CH in five-ring in **28**), 143.27 (2 C_q in *rac*-**27**), 138.93 (2 olefin. CH in chain in **28**), 138.85 (2 olefin. CH in

chain in *rac*-**27**), 138.80 (2 olefin. CH in chain in *rac*-**27**), 133.18 (2 olefin. CH in five-ring in *rac*-**27**), 129.98 (olefin. CH in five-ring in **28**), 126.47 (arom. CH in **28**), 126.32 (2 arom. CH in *rac*-**27**), 124.94 (arom. CH in **28**), 124.72 (2 arom. CH in *rac*-**27**), 122.94 (2 arom. CH in *rac*-**27**), 121.59 (arom. CH in **28**), 121.12 (arom. CH in **28**), 119.10 (2 arom. CH in *rac*-**27**), 114.86 (2 olefin. CH_2 in chain in *rac*-**27**), 114.67 (2 olefin. CH_2 in chain in *rac*-**27**), 114.50 (2 olefin. CH_2 in chain in **28**), 57.53 (C_q in **28**), 48.93 (2 aliph. CH in five-ring in *rac*-**27**), 37.07 (2 aliph. CH_2 in chain in **28**), 34.43 (2 aliph. CH_2 in chain in **28**), 34.19 (2 aliph. CH_2 in chain in *rac*-**27**), 33.76 (2 aliph. CH_2 in chain in *rac*-**27**), 31.41 (2 aliph. CH_2 in chain in *rac*-**27**), 27.32 (2 aliph. CH_2 in chain in *rac*-**27**), 27.14 (2 aliph. CH_2 in chain in *rac*-**27**), 26.99 (2 aliph. CH_2 in chain in *rac*-**27**), 23.76 (2 aliph. CH_2 in chain in **28**) ppm. EIMS (70eV): calcd. $C_{19}H_{24}$ 252.1878; found 252.1881.

A Mixture of Allyl(dimethyl)(3-pent-4-enyl-3*H*-inden-1-yl)silane (*rac*-29), Allyl(dimethyl)(3-pent-4-enyl-1*H*-inden-1-yl)silane (*rac*-30) and Allyl(dimethyl)(1-pent-4-enyl-1*H*-inden-1-yl)silane (*rac*-31): By applying the General Procedure 4, indene (420 μL, 3.6 mmol), *n*BuLi (2 × 1.5 mL, 2×4.7 mmol, 2.5 M in *n*-hexane), allyl(chloro) dimethylsilane (600 μL, 4.0 mmol) and 5-bromo-1-pentene (473 μL, 4.0 mmol) gave 0.7592 g (74%) of a mixture of the title compounds as a yellow oil. The ratio between the obtained compounds was 1:0.8:0.08. ^{1}H NMR (600 MHz, 25 °C, $CDCl_3$): δ = 7.51 (m, 2 H, 2 arom. CH in *rac*-**31**), 7.46 (m, 8 H, 4 arom. CH in *rac*-**29** and 4 arom. CH in *rac*-**30**), 7.38 (m, 2 H, 2 arom. CH in *rac*-**31**), 7.30 (m, 6 H, 2 arom. CH in *rac*-**29**, 2 arom. CH in *rac*-**30** and 2 arom. CH in *rac*-**31**), 7.22 (m, 6 H, 2 arom. CH in *rac*-**29**, 2 arom. CH in *rac*-**30** and 2 arom. CH in *rac*-**31**), 6.84 (d, J = 5.4 Hz, 2 H, 2 olefin. CH in five-ring in *rac*-**31**), 6.80 (m, 2 H, 2 olefin. CH in five-ring in *rac*-**29**), 6.58 (d, J = 5.4 Hz, 2 H, 2 olefin. CH in five-ring in *rac*-**31**), 6.35 (m, 2 H, 2 olefin. CH in five-ring in *rac*-**30**), 5.93 (m, 2 H, 2 olefin. CH in chain in *rac*-**30**), 5.84 (m, 4 H, 4 olefin. CH in chain in *rac*-**29**), 5.74 (m, 2 H, 2 olefin. CH in chain in *rac*-**30**), 5.35 (a X-part of an ABX – system, m, 4 H, 4 olefin. CH in chain in *rac*-**31**)*, 4.95 (m, 20 H, 4 olefin. CH_2 in chain in *rac*-**29**, 4 olefin. CH_2 in chain in *rac*-**30** and 2 olefin. CH_2 in chain in *rac*-**31**), 4.75 (m, 4 H, 2 olefin. CH_2 in chain in *rac*-**31**), 3.49 (m, 4 H, 2 aliph. CH in five-ring in *rac*-**29** and 2 aliph. CH in five-ring in *rac*-**30**), 2.75 (t, J = 7.7 Hz, 4 H, 2 aliph. CH_2 in chain in *rac*-**31**), 2.67 (t, J = 7.5 Hz, 4 H, 2 aliph. CH_2 in chain in *rac*-**30**), 2.22 (m, 4 H, 2 aliph. CH_2 in chain in *rac*-**30**), 2.12 (m, 4 H, 2 aliph. CH_2 in chain in *rac*-**29**), 1.93 (m, 2 H, 2 aliph. CH in chain in *rac*-**29**), 1.84 (m, 12 H, 2 aliph. CH_2 in chain in *rac*-**29**, 2 aliph. CH_2 in chain in *rac*-**30** and 2 aliph. CH_2 in chain in *rac*-**31**), 1.56 (m, 8 H, 2 aliph. CH_2 in chain in *rac*-**29** and 2 aliph. CH_2 in chain in *rac*-**30**), 1.49 (m, 2 H, 2 aliph. CH in chain in *rac*-**29**), 1.35 (m, 2 H, 2 aliph. CH in chain in *rac*-**31**), 1.31–1.26 (A-part of an ABX – system, dd, J_{AB} = –13.5 Hz, J_{AAX} = 8.2 Hz, 2 H, 2 aliph. CH in chain in *rac*-**31**)*, 1.22–1.17 (a B-part of an ABX – system, dd, J_{AB} = –13.5 Hz, J_{AAX} = 8.2 Hz, 2 H, 2 aliph. CH in chain in *rac*-**31**)*, 1.16 (m, 2 H, 2 aliph. CH in chain in *rac*-**31**), 0.35 (s, 12 H, 4 CH_3 in *rac*-**29**), 0.07 (s, 6 H, 2 CH_3 in *rac*-**31**), 0.02 (s, 6 H, 2 CH_3 in *rac*-**31**), –0.01 (s, 6 H, 2 CH_3 in *rac*-**30**), –0.08 (s, 6 H, 2 CH_3 in *rac*-**30**) ppm. ^{13}C NMR (150.9 MHz, 25 °C, $CDCl_3$): δ = 150.35 (2 olefin. CH in five-ring in *rac*-**29**), 148.77 (2 C_q in *rac*-**29**), 147.93 (2 C_q in *rac*-**31**), 147.54 (2 C_q in *rac*-**29**), 146.04 (2 C_q in *rac*-**30**), 144.71 (2 C_q in *rac*-**30**), 144.44 (2 C_q in *rac*-**31**), 142.20 (2 C_q in *rac*-**29**), 141.99 (2 C_q in *rac*-**30**), 139.39 (2 olefin. CH in chain in *rac*-**31**), 138.84 (2 olefin. CH in chain in *rac*-**29** or *rac*-**30**), 138.74 (2 olefin. CH in chain in *rac*-**29** or *rac*-**30**), 135.01 (2 olefin. CH in chain in *rac*-**31**), 134.83 (2 olefin. CH in chain in *rac*-**29**), 134.53 (2 olefin. CH in chain in *rac*-**30**), 131.63 (2 olefin. CH in five-ring in *rac*-**31**), 131.00 (2 olefin. CH in five-ring in *rac*-**30**), 129.56 (2 olefin. CH in five-ring in *rac*-**30**), 126.54 (2 arom. CH in *rac*-**31**), 126.43 (2 arom. CH in *rac*-**29** or *rac*-**30**), 124.75 (2 arom. CH in *rac*-**29** or *rac*-**30**), 124.53 (2 arom. CH in *rac*-**29** or *rac*-**30**), 124.08 (2 arom. CH in *rac*-**31**), 123.79 (2 arom. CH in *rac*-**29** or *rac*-**30**), 123.10 (2 arom. CH in *rac*-**29** or *rac*-**30**), 123.00 (2 arom. CH in *rac*-**29** or *rac*-**30**), 122.96 (2 arom. CH in *rac*-**31**), 122.19 (2 arom. CH in *rac*-**29** or *rac*-**30**), 121.11 (2 arom. CH in *rac*-**31**), 119.21 (2 arom. CH in *rac*-**29** or *rac*-**30**), 144.85 (2 olefin. CH_2 in chain in *rac*-**29** or *rac*-**30**), 144.75 (2 olefin. CH_2 in chain in *rac*-**29** or *rac*-**30**), 144.45 (2 olefin. CH_2 in chain in *rac*-**31**), 113.64 (2 olefin. CH_2 in chain in *rac*-**29** or *rac*-**30**), 113.48 (2 olefin. CH_2 in chain in *rac*-**29** or *rac*-**30**), 113.27 (2 olefin. CH_2 in chain in *rac*-**31**), 58.62 (2 C_q in *rac*-**31**), 51.92 (2 aliph. CH in five-ring in *rac*-**29**), 43.24 (2 aliph. CH in five-ring in *rac*-**30**), 37.08 (2 aliph. CH_2 in chain in *rac*-**31**), 34.17 (2 aliph. CH_2 in chain in *rac*-**29**), 33.88 (2 aliph. CH_2 in chain in *rac*-**30**), 31.05 (2 aliph. CH_2 in chain in *rac*-**29**), 30.33 (2 aliph. CH_2 in chain in *rac*-**31**), 28.61 (2 aliph. CH_2 in chain in *rac*-**31**), 28.11 (2 aliph. CH_2 in chain in *rac*-**30**), 27.21 (2 aliph. CH_2 in chain in *rac*-**30**), 26.81 (2 aliph. CH_2 in chain in *rac*-**29**), 23.56 (2 aliph. CH_2 in chain in *rac*-**29**), 22.57 (2 aliph. CH_2 in chain in *rac*-**30**), 22.25 (2 aliph. CH_2 in chain in *rac*-**31**), –2.48 (2 CH_3 in *rac*-**31**), –2.99 (2 CH_3 in *rac*-**31**), –3.13 (2 CH_3 in *rac*-**29**), –3.18 (2 CH_3 in *rac*-**29**), –4.34 (2 CH_3 in *rac*-**30**), –4.73 (2 CH_3 in *rac*-**30**) ppm. ^{29}Si NMR (119.3 MHz, 25 °C, $CDCl_3$): δ = 3.39 (2 Si in *rac*-**30**), 0.78 (2 Si in *rac*-**31**), –9.68 (2 Si in *rac*-**29**) ppm. EIMS (70eV): calcd. $C_{19}H_{26}Si$ 282.1804; found 282.1811.

A Mixture of 4,4a-Dihydro-1*H*-fluorene (*rac*-4) and 4,9-Dihydro-1*H*-fluorene (5). Ring-Closing Metathesis with Grubbs 1st Generation Catalyst: To a solution of 1,2-diallyl-1*H*-indene (*rac*-**1**), [(1*E*,4*E*)-2-hexa-1,4-dienyl]-1*H*-indene (**2**) and (*E*/*Z*)-1-allyl-2-(prop-2-enylidene)indane (*rac*-**3**) (0.3465 g, 1.8 mmol) in dry dichloromethane (5 mL) was added Grubbs 1st generation catalyst (0.1534 g, 0.2 mmol) in portions. The resulting reaction mixture was refluxed for 3.5 h before the solvent was removed under reduced pressure. The residue was directly chromatographed on silica gel (hexane as eluent) and 0.2049 g (69%) of a 8:1 mixture of the title compounds as a colorless oil. EIMS (70eV): calcd. $C_{13}H_{12}$ 168.0939; found 168.0931. The ^{1}H NMR spectroscopic data (600 MHz, 25 °C, $CDCl_3$) and the ^{13}C NMR spectroscopic data (150.9 MHz, 25 °C, $CDCl_3$) are analogous to those reported in the context of the experiment with the 2nd generation catalyst (vide supra).

Supporting Information (for details see the footnote on the first page of this article): Copies of ^{13}C NMR spectra of the compounds reported are provided.

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