Cycloaddition of diazoketones to [60]fullerene in the presence of the catalytic system $Pd(acac)_2$—PPh_3—Et_3Al

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A method for the selective and efficient synthesis of methanofullerenes by cycloaddition of diazoketones to [60]fullerene in the presence of a three-component catalytic system $Pd(acac)_2$—PPh_3—Et_3Al has been developed.

Key words: metal complex catalysis, cycloaddition, [60]fullerene, diazoketones, methanofullerenes.

Functionally substituted fullerenes are of particular interest and practical value. A great number of publications is devoted to the synthesis and physicochemical properties of these compounds.[1—7] The main part of these works is focused on the synthesis and study of the properties of homo- and methanofullerenes, which synthesized by the Bingel reaction[8] as well as by thermal[9] or catalytical[10] cycloaddition of diazo compounds to [60]fullerene. The known synthetic methods toward cyclopropafullerenes by reaction of [60]fullerene with diazo compounds, including diazoketones,[11—14] have little preparative significance due to low yields of the target methanofullerenes or require stoichiometric amounts of such expensive promoters as ruthenium complexes.[9,10]

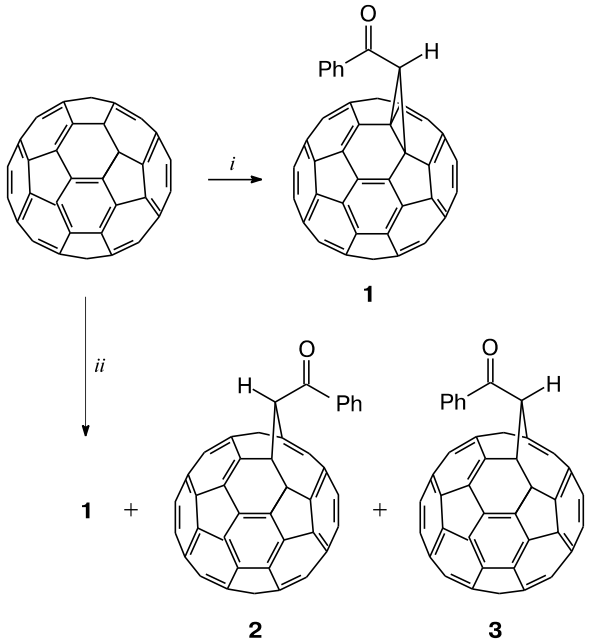
Recently,[15—18] we have reported the synthesis of homo- and methanofullerenes in high yields by cycloaddition of diazo compounds (diazomethane,[15] diazoalkanes,[16] and diazoacetates[17,18]) to [60]fullerene in the presence of palladium complexes. In continuation of this research, in the present work we studied cycloaddition of diazoketones to [60]fullerene in the presence of palladium complexes. The effect of the substituent in the starting diazoketone on the selectivity of the reaction and the yields of the target functionalized methanofullerenes have also been studied.

Results and Discussion

Among a number of tested transition metal complexes, a three-component catalyst prepared from $Pd(acac)_2$, PPh_3, and Et_3Al (1 : 2 : 4) was found most efficient and selective in the reactions under study and in the reactions of [60]fullerene with diazoacetates as well. In this regard, all subsequent experiments were carried out using this catalytic system. Cycloaddition of 2-oxo-2-phenyldiazoethane to [60]fullerene was used as a model reaction. It is known that the reaction of 2-oxo-2-phenyldiazoethane with [60]fullerene at ~20 °C for 1 week resulted in a mixture of cycloadducts **1—3** in the yield of ~30% (**1** : **2** : **3** = 1 : 1 : 3).[14]

We found that the reaction of 2-oxo-2-phenyldiazoethane with [60]fullerene (reactant ratio 5 : 1, respectively) in the presence of a three-component catalytic system $Pd(acac)_2$—PPh_3—Et_3Al (1 : 2 : 4, 20 mol.%) at 80 °C (1 h, *o*-dichlorobenzene) furnished exclusively 6,6-closed cycloadduct **1** in ~40% yield (Scheme 1). No increase in the

Scheme 1



i. $N_2CHC(O)Ph$, $Pd(acac)_2$—PPh_3—Et_3Al (1 : 2 : 4); *ii*. $N_2CHC(O)Ph$

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total yield (~43%) was observed when the reaction time was prolonged up to 2 h. When the reaction under study was carried out under the same conditions but without catalytic system, a mixture of 6,6-closed (**1**) and 5,6-open (**2**, **3**) adducts was obtained in 1 : 1 : 3 ratio in ~35% total yield. No formation of bis-adducts and dihydrofuranofullerene, which formed usually in the thermal reaction of [60]fullerene with diazoketones,[11—13] were observed in our experiments.

The ^{1}H NMR spectrum of compound **1** exhibited the singlet signal (δ_H 5.65), which was attributed to the proton at the bridging C atom, doublet signal (δ_H 8.48 (J = 7.6 Hz)) and two triplet signals (δ_H 7.70 (J = 7.6 Hz) and 7.72 (J = 7.6 Hz)) characteristic of the protons of the phenyl group. These spectral data are in good accord with previously published data[14] on the cycloaddition of 2-oxo-2-phenyldiazoethane to [60]fullerene and confirmed the formation of methanofullerene **1**.

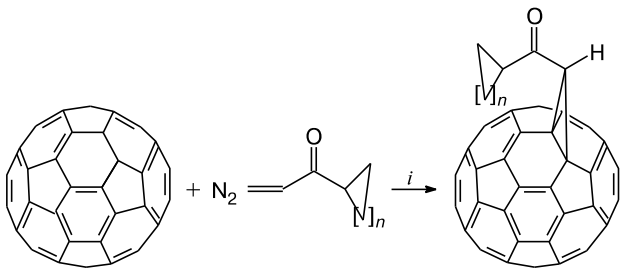
The data obtained from the ^{1}H and ^{13}C NMR spectra and 2D experiments (HHCOSY, HSQC, HMBC) of a mixture of compounds **1—3** indicated that the studied reaction carried out under described conditions without catalyst resulted in 6,6-closed (**1**) and stereoisomeric 5,6-open (**2**, **3**) monoadducts.

In accordance with the ^{1}H NMR spectral data, 5,6-open adduct **3** was the main component in the mixture of compounds **1—3** (signal ratio 1 : 1 : 3). The singlet signal for the proton at the bridging C atom of compound **3** is shifted to the higher filed (δ_H 4.53) than those of stereoisomer **2** (δ_H 8.14) and methanofullerene **1** (δ_H 5.65). The ^{1}H and ^{13}C NMR spectral data of compounds **1—3** are consistent with the data documented.[14]

On the next step of our research, we extended the developed conditions (80 °C, 1 h, *o*-dichlorobenzene, a three-component catalytic system $Pd(acac)_2$—PPh_3—Et_3Al (1 : 2 : 4, 20 mol.%)) on the reactions of 2-oxo-2-cyclopropyl-, 2-oxo-2-cyclobutyl-, and 2-oxo-2-cyclopentyldiazoethanes with [60]fullerene. The corresponding methanofullerenes **4—6** were obtained in ~45% total yield (Scheme 2). No effects of the size of the carbocycle proximal to the carbonyl C atom in the starting diazo compound on the direction of the reaction and the yields of cycloadducts were found.

Selective formation of methanofullerenes **4—6** in the reactions of [60]fullerene with the corresponding diazoketones was confirmed by the ^{1}H NMR spectra and HMBC experiments. Thus, the protons at the bridging C atom exhibited the singlet signals in the range of δ 5; in the HMBC experiments, these signals gave cross peaks with the sp^3-hybridized C atoms of the fullerene core in the range of δ 72. The UV spectra of the individual compounds **4—6** exhibited narrow absorption band of moderate intensity in the range of 430 nm, which is fairly simple and reliable demonstration of the formation of 6,6-closed adducts. The mass spectra (MALDI—TOF) of methanofullerenes **4—6** revealed intensive peaks of molecular ions with m/z 802.734, 816.734, and 830.824, respectively, which also confirmed the formation of monoadducts.

Scheme 2



4: n = 1;
5: n = 2;
6: n = 3

i. $Pd(acac)_2$—PPh_3—Et_3Al (1 : 2 : 4), 80 °C, 1 h.

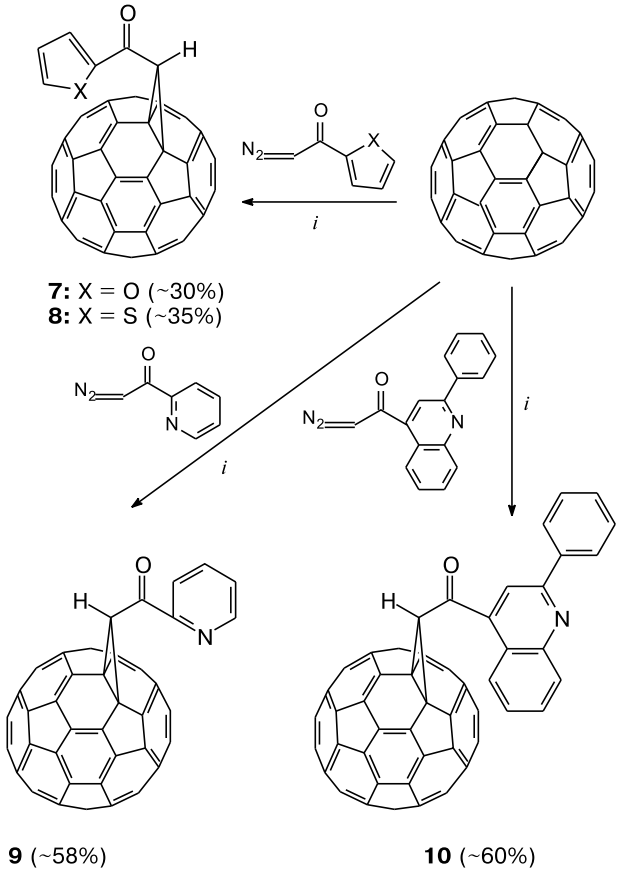
With the aim to extend the applicability of the reaction under investigation and study the activity of the complex catalytic system as well, we involved diazoacetyl-substituted O-, S-, and N-heterocycles in the cycloaddition with [60]fullerene. It was found that the reaction of [60]fullerene with 2-oxo-2-(furan-2-yl)-, 2-oxo-2-(thiophen-2-yl)-, 2-oxo-2-(pyridin-2-yl)-, and 2-oxo-2-(phenylquinolin-4-yl)diazoethanes (80 °C, 1 h) in the presence of $Pd(acac)_2$—PPh_3—Et_3Al (1 : 2 : 4, 20 mol.%) resulted in 6,6-closed cycloadducts **7—10** in the yields of 30—60% (Scheme 3). The N-heterocyclic diazoketones gave the highest yields of the target products (~60%).

The structures of the resulting compounds **7—10** were confirmed by the NMR, IR, and UV spectroscopy and the high resolution mass spectrometry.

Thus, the structure of cycloadduct **7** was confirmed by the 1D and 2D NMR spectroscopy. The three-spin system of the furan ring exhibited the doublet signal at δ_H 7.63 (^3J = 3.5 Hz), the doublet-of-doublet signal at δ_H 6.81 (^3J = 3.5 Hz, ^3J = 1.7 Hz), and the triplet signal at δ_H 7.86 with the line half-width measured at half-height ($\Delta w_{1/2}$) equal to 4.3 Hz. In the shift-correlated HSQC spectra, these signals exhibited the cross-peaks with the C atoms at δ_C 118.43, 113.46, and 147.10, respectively. In the HMBC experiment, the cross-peaks of the proton at the bridging C atom (δ_H 5.65) with the quaternary C atom of the furan ring (δ_C 148.65) and the sp^3-hybridized C atoms of the fullerene core (δ_C 72.09) were observed, which confirmed the formation of 6,6-closed adduct. In the ^{13}C NMR spectra, low intensity signal for the carbonyl group appeared at δ_C 190.10.

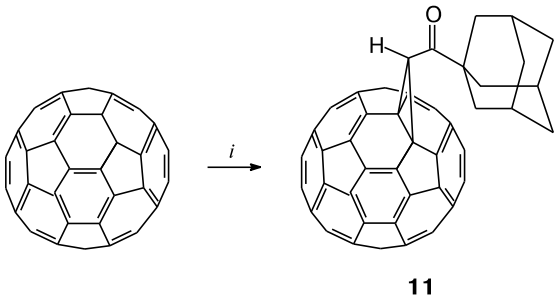
It was found that under developed conditions (80 °C, 1 h, $Pd(acac)_2$: PPh_3 : Et_3Al (1 : 2 : 4, 20 mol.%)), 2-oxo-2-(adamantan-1-yl)diazoethane reacted very selectively with [60]fullerene giving monoadduct **11** in ~40% yield (Scheme 4). In these experiments, no formation of the regioisomeric bis-adducts were observed.

Scheme 3



i. $Pd(acac)_2$—PPh_3—Et_3Al (1 : 2 : 4)

Scheme 4



i. $N_2CHCOAd$, $Pd(acac)_2$—PPh_3—Et_3Al (1 : 2 : 4), 80 °C, 1 h.

The exclusive formation of 6,6-closed cycloadduct **11** was confirmed based on the ^1H and ^{13}C NMR and 2D NMR experiments (HHCOSY, HSQC, HMBC) of **11**.

In summary, it was shown that cycloaddition of 2-oxo-2-aryl-, 2-oxo-2-cycloalkyl-, and 2-oxo-2-heteroaryldiazoethanes to [60]fullerene in the presence of a three-component catalytic system $Pd(acac)_2$—PPh_3—Et_3Al readily gives 6,6-closed cycloadducts in relatively high yields. No influence of the structure of the substituent at the carbonyl C atom on the direction of the reaction was found.

Experimantal

Commercially available [60]fullerene (99.5% pure, G. A. Razuvaev Institute of Organometallic Chemistry, Russian Academy of Sciences, Nizhniy Novgorod) was used. The reaction products were analyzed using an HPLC chromatograph Altex (model 330) (USA) equipped with an UV detector (340 nm). The mixtures were separated on a metal half-preparative column Cosmosil Buckyprep Waters (250×10 mm) at ~20 °C. Toluene was used as the eluent, the flow rate was 2.0 mL min^{-1}. The IR spectra were registered on a Specord 75 IR (Carl Zeiss Jena) spectrophotometer in KBr pellets. The UV spectra were recorded on Specord M-40 and Specord M-80 instruments in $CHCl_3$. The ^1H and ^{13}C NMR spectra were run on a Bruker Avance-400 spectrometer at 400.13 and 100.62 MHz, respectively. The mixture of $CDCl_3$ and CS_2 (1 : 5) was used as a solvent. The negative-ion mass spectra were obtained on a MALDI TOF/TOF Autoflex-III Bruker mass spectrometer without a matrix operating in a linear mode. Samples were dissolved in toluene prior to application on a metal target.

Cycloaddition of diazoketones to [60]fullerene (general procedure). A solutions of $Pd(acac)_2$ (0.00278 mmol) in *o*-dichlorobenzene (0.4 mL) and PPh_3 (0.00556 mmol) in *o*-dichlorobenzene (0.4 mL) were mixed in the glas flask, then a solution of Et_3Al (0.01112 mmol) in toluene (0.1 mL) was added with stirring under stream of nitrogen at –5 °C. The color of the solution changed from light yellow to light brown. A solution of [60]-fullerene (0.0139 mmol) in *o*-dichlorobenzene (0.1 mL) was added to the resulting catalyst at room temperature, the color of the solution turned dark green. The mixture was heated to 80 °C and a solution of the corresponding diazoketone (0.0695 mmol) in CH_2Cl_2 (0.5 mL) was added dropwise over a period of 5 min. The reaction mixture was stirred for 1 h at the corresponding temperature, then cooled to room temperature, treated with aqueous HCl, and extracted with toluene (7 mL). The organic layer was passed through short column filled with silica gel. The products **1—11** and [60]fullerene were separated by preparative HPLC using toluene as the eluent. The structures of compounds **1—3** were established by comparing the spectral data with those published previously.[14]

3′*H*-Cyclopropa[1,9](C_{60}-I_h)[5,6]fulleren-3′-yl(cyclopropyl)methanone (4). IR, ν/cm^{-1}: 540; 820; 1040; 1110; 1270; 1660. UV ($CHCl_3$), λ_{max}/nm: 260, 327, 424. ^1H NMR, δ: 1.15—1.40 (m, 4 H, 2 CH_2,); 2.62—2.71 (m, 1 H, CH); 5.19 (s, 1 H, CH). ^{13}C NMR, δ: 12.99; 22.66; 46.60; 72.42 (sp^3); 138.17; 138.35; 140.29; 141.23; 142.08; 142.14; 142.24; 142.57; 142.81; 143.02; 143.12; 143.46; 143.75; 143.99; 144.37; 144.60; 144.70; 144.83; 145.20; 145.23; 145.25; 145.51; 146.31; 148.24, 190.02 (CO). MS (MALDI—TOF), *m/z*: [M^+], found 802.734, calculated 802.743, $C_{65}H_6O$.

Cyclobutyl-3′*H*-cyclopropa[1,9](C_{60}-I_h)[5,6]fulleren-3′-ylmethanone (5). IR, ν/cm^{-1}: 520; 1080; 1120; 1200; 1660. UV ($CHCl_3$), λ_{max}/nm: 260, 330, 427. ^1H NMR, δ: 2.00—2.80 (m, 6 H, CH_2); 3.96—4.09 (m, 1 H, CH); 4.94 (s, 1 H, CH). ^{13}C NMR, δ: 18.73; 25.08; 43.86; 47.52; 72.07; 139.70; 140.47; 140.99; 141.23; 142.03; 142.13; 142.31; 142.48; 142.78; 143.03; 143.40; 143.71; 143.78; 144.01; 144.11; 144.39; 144.59; 144.67; 145.06; 145.17; 145.45; 145.51; 145.60; 146.38; 147.99; 154.52; 155.34; 190.06 (CO). MS (MALDI—TOF), *m/z*: [M^+], found 816.734, calculated 816.769, $C_{66}H_8O$.

Cyclopentyl-3´*H*-cyclopropa[1,9](C_{60}-I_h)[5,6]fulleren-3´-ylmethanone (6). IR, ν/cm^{-1}: 530; 1130; 1180; 1200; 1230; 1670. UV ($CHCl_3$), λ_{max}/nm: 261, 327, 425. ^{1}H NMR, δ: 1.50—2.35 (m, 8 H, CH_2); 3.30—3.45 (m, 1 H, CH); 5.06 (s, 1 H, CH). ^{13}C NMR, δ: 26.65; 29.29; 45.29; 47.39; 72.39; 136.39; 138.20; 140.19; 140.51; 140.60; 140.89; 140.98; 141.23; 141.80; 142.04; 142.12; 142.33; 142.50; 142.80; 143.01; 143.22; 143.43; 143.71; 144.05; 144.59; 144.70; 145.07; 145.17; 145.51; 146.17; 146.48; 147.10; 148.29; 190.53 (CO). MS (MALDI—TOF), *m/z*: [M$^+$], found 830.812, calculated 830.796, $C_{67}H_{10}O$.

3´*H*-Cyclopropa[1,9](C_{60}-I_h)[5,6]fulleren-3´-yl(furan-2-yl)methanone (7). IR, ν/cm^{-1}: 520; 780; 1220; 1380; 1460; 1670. UV ($CHCl_3$), λ_{max}/nm: 260, 333, 430. ^{1}H NMR, δ: 5.65 (s, 1 H, CH); 6.81 (dd, 1 H, CH, ^3J = 3.5 Hz, ^3J = 1.7 Hz); 7.63 (d, 1 H, CH, ^3J = 3.5 Hz); 7.86 (t, 1 H, CH, $\Delta w_{1/2}$ = 4.3 Hz). ^{13}C NMR, δ: 42.45; 72.09; 113.46; 118.43; 140.87; 140.97; 141.25; 142.00; 142.20; 142.35; 142.66; 142.83; 143.03; 143.08; 143.34; 143.80; 144.05; 144.44; 144.63; 144.69; 144.95; 145.07; 145.19; 145.22; 145.70; 146.35; 147.10; 148.65; 148.97; 149.51; 152.89; 190.10 (CO). MS (MALDI—TOF), *m/z*: [M$^+$], found 828.724, calculated 828.737, $C_{66}H_4O_2$.

3´*H*-Cyclopropa[1,9](C_{60}-I_h)[5,6]fulleren-3´-yl(thiophen-2-yl)methanone (8). IR, ν/cm^{-1}: 540; 750; 1220; 1380; 1480; 1660. UV ($CHCl_3$), λ_{max}/nm: 261, 326, 425. ^{1}H NMR, δ: 5.54 (s, 1 H, CH); 7.39 (t, 1 H, CH, *J* = 4.4 Hz); 7.90 (d, 1 H, CH, *J* = 3.2 Hz); 8.32 (d, 1 H, CH, *J* = 4.4 Hz). ^{13}C NMR, δ: 44.06; 72.11; 128.83; 132.98; 135.36; 140.27; 141.06; 141.31; 142.13; 142.17; 142.35; 142.50; 142.83; 142.96; 143.04; 143.10; 143.19; 143.22; 143.40; 143.46; 143.75; 144.04; 144.47; 144.69; 144.77; 144.95; 145.10; 145.26; 145.33; 145.53; 146.20; 190.56 (CO). MS (MALDI—TOF), *m/z*: [M$^+$], found 844.744, calculated 844.803, $C_{66}H_4OS$.

3´*H*-Cyclopropa[1,9](C_{60}-I_h)[5,6]fulleren-3´-yl(pyridin-2-yl)methanone (9). IR, ν/cm^{-1}: 580; 800; 1250; 1470; 1670. UV ($CHCl_3$), λ_{max}/nm: 260, 330, 427. ^{1}H NMR, δ: 6.62 (s, 1 H, CH); 7.68 (t, 1 H, CH, *J* = 7.2 Hz); 8.06 (t, 1 H, CH, *J* = 7.2 Hz); 8.34 (d, 1 H, CH, *J* = 7.6 Hz); 8.95 (d, 1 H, CH, *J* = 7.6 Hz). ^{13}C NMR, δ: 41.45; 73.25; 122.64; 127.86; 137.21; 140.89; 140.96; 141.24; 141.99; 142.19; 142.35; 142.65; 142.85; 143.03; 143.07; 143.38; 143.79; 144.05; 144.41; 144.63; 144.69; 144.95; 145.05; 145.19; 145.26; 145.69; 146.30; 148.97; 149.51; 152.89; 192.02 (CO). MS (MALDI—TOF), *m/z*: [M$^+$], found 839.724, calculated 839.763, $C_{67}H_5NO$.

3´*H*-Cyclopropa[1,9](C_{60}-I_h)[5,6]fulleren-3´-yl(2-phenylquinolin-4-yl)methanone (10). IR, ν/cm^{-1}: 520; 1180; 1210; 1430; 1630. UV ($CHCl_3$), λ_{max}/nm: 260, 329, 425. ^{1}H NMR, δ: 5.67 (s, 1 H, CH); 7.45 (t, 1 H, CH, Ph, *J* = 7.2 Hz); 7.58 (t, 2 H, CH, Ph, *J* = 7.2 Hz); 7.87 (t, 1 H, CH, C_9H_5N, *J* = 7.2 Hz); 8.32 (d, 2 H, CH, Ph, *J* = 7.2 Hz); 8.30—8.38 (m, 2 H, CH, C_9H_5N); 8.75 (s, 1 H, CH, C_9H_5N); 8.80 (d, 1 H, CH, C_9H_5N, *J* = 7.2 Hz). ^{13}C NMR, δ: 45.74; 72.44; 118.47; 125.38; 128.70; 129.21; 130.17; 130.58; 132.08; 132.18; 136.79; 138.55; 140.60; 141.14; 141.38; 142.05; 142.13; 142.31; 142.45; 142.85; 143.06; 143.23; 143.42; 143.75; 144.03; 144.28; 144.57; 144.75; 145.11; 145.28; 145.34; 145.40; 145.61; 146.24; 146.38; 146.65; 147.60; 149.66; 156.72; 196.55 (CO). MS (MALDI—TOF), *m/z*: [M$^+$], found 965.858, calculated 965.917, $C_{77}H_{11}NO$.

(Adamantan-1-yl)-3´*H*-cyclopropa[1,9](C_{60}-I_h)[5,6]fulleren-3´-ylmethanone (11). IR, ν/cm^{-1}: 520; 800; 1250; 1430; 1670. UV ($CHCl_3$), λ_{max}/nm: 260, 320, 427. ^{1}H NMR, δ: 1.40—2.00 (m, 15 H, CH_2 and CH); 5.26 (s, 1 H, CH). ^{13}C NMR, δ: 28.50; 36.85; 38.61; 42.29; 47.45; 72.10 (sp^3); 138.09; 140.24; 140.97; 141.22; 142.10; 142.30; 142.77; 142.98; 143.17; 143.39; 143.71; 144.00; 144.34; 144.61; 145.08; 145.18; 145.29; 145.45; 145.97; 146.71; 148.36; 196.30 (CO). MS (MALDI—TOF), *m/z*: [M$^+$], found 896.848, calculated 896.897, $C_{72}H_{16}O$.

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