Journal of the Serbian Chemical Society
JSCS@tmf.bg.ac.rs • www.shd.org.rs/JSCS
J. Serb. Chem. Soc. 78 (6) S62–S74 (2013) *Supplementary material*

SUPPLEMENTARY MATERIAL TO

Fe_3O_4 nanoparticles: a highly efficient and easily reusable catalyst for the one-pot synthesis of xanthene derivatives under solvent-free conditions

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J. Serb. Chem. Soc. 78 (6) (2013) 769–779

SPECTRAL DATA FOR KNOWN COMPOUNDS

*14-Phenyl-14*H*-dibenzo[*a,j*]xanthene (**4a**).* White crystals; yield: 88 %; m.p.: 181–183 °C; Anal. Calcd. for $C_{27}H_{18}O$ (FW: 358.14): C, 90.47; H 5.06 %. Found: C, 90.22; H, 5.21 %; FT-IR (KBr, cm^{-1}): 3052 (=C–H stretching of aromatic ring), 1625 (C=C– stretching of aromatic ring), 1575, 1253 (C–O stretching), 814; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 6.44 (1H, *s*, CH), 7.16 (2H, *d*, *J* = 8.0 Hz, Ar-H), 7.25–7.33 (4H, *m*, Ar-H), 7.37–7.41 (5H, *m*, Ar-H), 7.46 (4H, *t*, *J* = 7.8 Hz, Ar-H), 7.72 (2H, *d*, *J* = 8.0 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 31.9 (CH), 115.8,116.5, 119.2, 123.1, 124.9, 126.6, 128.2, 129.5, 130.2, 132.2, 136.9, 141.5, 148.1, 149.9 (Ar–C).

*14-(4-Methoxyphenyl)-14*H*-dibenzo[*a,j*]xanthene (**4b**).* Yellow crystals; yield: 85 %; m.p.: 202–204 °C; Anal. Calcd. for $C_{28}H_{20}O_2$ (FW: 388.15): C, 86.57; H, 5.19 %. Found: C, 86.72; H, 5.06 %; FT-IR (KBr, cm^{-1}): 3042 (=C–H stretching of aromatic ring), 1625 (C=C– stretching of aromatic ring), 1594, 1251 (C–O stretching), 1236 (C–O stretching), 811; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / / ppm): 3.62 (3H, *s*, OCH_3), 6.45 (1H, *s*, CH), 6.68 (2H, *d*, *J* = 7.9 Hz, Ar-H), 7.27–7.40 (4H, *m*, Ar-H), 7.42–7.46 (4H, *m*, Ar-H), 7.79 (2H, *d*, *J* = 8.0 Hz, Ar-H), 8.83 (2H, *d*, *J* = 8.0 Hz, Ar-H), 8.39 (2H, *d*, *J* = 7.9 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 37.1 (CH), 55.1 (OCH_3), 113.8, 117.5, 118.0, 122.7, 124.7, 126.7, 128.7, 128.8, 129.2, 131.1, 131.4, 137.4, 148.6, 157.8 (Ar–C).

*14-(4-Chlorophenyl)-14*H*-dibenzo[*a,j*]xanthene (**4c**).* Yellow crystals; yield: 90 %; m.p.: 289–290 °C; Anal. Calcd. for $C_{27}H_{17}ClO$ (FW: 392.10): C, 82.54; H, 4.36 %. Found: C, 86.27; H, 4.52 %; FT-IR (KBr, cm^{-1}): 3044 (=C–H

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stretching of aromatic ring), 1621 (C=C– stretching of aromatic ring), 1588, 1246 (C–O stretching), 814; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 6.46 (1H, *s*, CH), 7.11 (2H, *d*, *J* = 8.1 Hz, Ar-H), 7.26–7.45 (6H, *m*, Ar-H), 7.48 (2H, *t*, *J* = 7.6 Hz, Ar-H), 7.57–7.82 (4H, *m*, Ar-H), 8.32 (2H, *d*, *J* = 8.1 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 36.9 (CH), 115.8, 116.9, 118.1, 121.3, 124.2, 125.8, 127.5, 128.1, 129.5, 131.3, 132.2, 136.3, 148.5, 158.7 (Ar–C).

*14-(4-Bromophenyl)-14*H-*dibenzo[*a,j*]xanthene* (***4d***). Yellow crystals; yield: 90 %; m.p.: 296–297 °C; Anal. Calcd. for $C_{27}H_{17}BrO$ (FW: 436.05): C, 74.15; H, 3.92 %. Found: C, 74.31; H, 3.71 %; FT-IR (KBr, cm^{-1}): 3042 (=C–H stretching of aromatic ring), 1624 (C=C– stretching of aromatic ring), 1521, 1244 (C–O stretching), 812; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 6.45 (1H, *s*, CH), 7.18 (2H, *d*, *J* = 8.1 Hz, Ar-H), 7.23–7.31 (4H, *m*, Ar-H), 7.40 (2H, *t*, *J* = 7.8 Hz, Ar-H), 7.44–7.49 (6H, *m*, Ar-H), 8.17 (2H, *d*, *J* = 8.1 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 37.1 (CH), 116.1, 116.9, 117.8, 121.3, 123.1, 125.4, 127.2, 127.9, 130.1, 132.2, 133.7, 136.9, 146.5, 155.9 (Ar–C).

*14-(4-Nitrophenyl)-14*H-*dibenzo[*a,j*]xanthene* (***4e***). Yellow crystals; Yield: 92 %; m.p.: 311–312 °C; Anal. Calcd. for $C_{27}H_{17}NO_3$ (FW: 403.12): C, 80.38; H, 4.25; N, 3.47 %. Found: C, 80.54; H, 4.31; N, 3.28 %; FT-IR (KBr, cm^{-1}): 3041 (=C–H stretching of aromatic ring), 1622 (C=C– stretching of aromatic ring), 1583, 1548 (NO_2), 1348 (NO_2), 1242 (C–O stretching), 809; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 6.61 (1H, *s*, CH), 7.44 (2H, *t*, *J* = 8.0 Hz, Ar-H), 7.52 (2H, *d*, *J* = 8.1 Hz, Ar-H), 7.60 (2H, *t*, *J* = 8.0 Hz, Ar-H), 7.69 (2H, *d*, *J* = = 8.1 Hz, Ar-H), 7.84–7.87 (4H, *m*, Ar-H), 8.01 (2H, *d*, *J* = 8.0 Hz, Ar-H), 8.29 (2H, *d*, *J* = 8.0 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 36.9 (CH), 115.9, 116.2, 117.1, 120.2, 122.5, 124.8, 126.1, 128.1, 129.7, 132.6, 134.1, 135.9, 148.3, 159.1 (Ar–C).

*14-(*p-*Tolyl)-14*H-*dibenzo[*a,j*]xanthene* (***4f***). Yellow crystals; yield: 86 %; m.p.: 228–230 °C; Anal. Calcd. for $C_{28}H_{20}O$ (FW: 372.15): C, 90.29; H, 5.41 %. Found: C, 90.52; H, 5.25 %; FT-IR (KBr, cm^{-1}): 3047 (=C–H stretching of aromatic ring), 1619 (C=C– stretching of aromatic ring), 1515, 1249 (C–O stretching), 809; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.16 (3H, *s*, CH_3), 6.46 (1H, *s*, CH), 6.96 (2H, *d*, *J* = 7.9 Hz, Ar-H), 7.39–7.43 (4H, *m*, Ar-H), 7.49 (2H, *d*, *J* = 8.1 Hz, Ar-H), 7.57 (2H, *d*, *J* = 7.9 Hz, Ar-H), 7.78–7.84 (4H, *m*, Ar-H), 8.41 (2H, *d*, *J* = 8.1 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 20.9 (CH_3), 37.7 (CH), 116.3, 117.5, 118.1, 122.8, 124.2, 126.8, 128.1, 128.8, 129.2, 131.1, 135.9, 142.2, 148.7, 149.8 (Ar–C).

*14-(2-Nitrophenyl)-14*H-*dibenzo[*a,j*]xanthene* (***4g***). White crystals; yield: 89 %; m.p.: 214–216 °C; Anal. Calcd. for $C_{27}H_{17}NO_3$ (FW: 403.12): C, 80.38; H, 4.25; N, 3.47 %. Found: C, 80.21; H, 4.33; N, 3.58 %; FT-IR (KBr, cm^{-1}): 3067 (=C–H stretching of aromatic ring), 2925, 1548 (NO_2), 1481 (C=C– stretching of aromatic ring), 1357 (NO_2), 1243 (C–O stretching), 749; ^{1}H-NMR (400 MHz,

$CDCl_3$, δ / ppm): 6.81 (1H, *s*, CH), 6.84–7.27 (4H, *m*, Ar-H), 7.41–7.50 (4H, *m*, Ar-H), 7.61 (2H, *t*, *J* = 7.9 Hz, Ar-H), 7.80–7.84 (4H, *m*, Ar-H), 8.42 (2H, *t*, *J* = = 7.8 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 30.2 (CH), 115.1, 115.2, 116.7, 117.9, 122.3, 124.4, 125.1, 127.1, 128.2, 128.6, 129.1, 130.9, 131.1, 131.5, 132.4 (Ar–C).

*14-(4-Fluorophenyl)-14*H-*dibenzo[*a,j*]xanthene* (***4h***). Yellow crystals; yield: 91 %; m.p.: 238–240 °C; Anal. Calcd. for $C_{27}H_{17}FO$ (FW: 376.13): C, 86.15; H, 4.55 %. Found: C, 86.28; H, 4.41 %; FT-IR (KBr, cm^{-1}): 3052 (=C–H stretching of aromatic ring), 1618 (C=C– stretching of aromatic ring), 1581, 1223 (C–O stretching), 812; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 6.41 (1H, *s*, CH), 7.07 (2H, *d*, *J* = 8.0 Hz, Ar-H), 7.17–7.24 (4H, *m*, Ar-H), 7.38 (2H, *d*, *J* = 7.9 Hz, Ar-H), 7.51–7.54 (6H, *m*, Ar-H), 8.12 (2H, *d*, *J* = 8.0 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 36.8 (CH), 115.8, 117.1, 117.7, 122.1, 124.5, 125.2, 127.7, 127.9, 131.6, 132.1, 133.8, 137.1, 146.5, 156.1 (Ar–C).

*14-(3-Chlorophenyl)-14*H-*dibenzo[*a,j*]xanthene* (***4i***). White crystals; yield: 85 %; m.p.: 211–212 °C; Anal. Calcd. for $C_{27}H_{17}ClO$ (FW: 392.10): C, 82.54; H, 4.36 %. Found: C, 82.41; H, 4.48 %; FT-IR (KBr, cm^{-1}): 3045 (=C–H stretching of aromatic ring), 1592, 1513 (C=C– stretching of aromatic ring), 1247 (C–O stretching), 808; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 6.46 (1H, *s*, CH), 7.12 (2H, *d*, *J* = 7.9 Hz, Ar-H), 7.26 (2H, *d*, *J* = 8.1 Hz, Ar-H), 7.54–7.62 (5H, *m*, Ar-H), 7.79–7.84 (4H, *m*, Ar-H), 8.22 (1H, *s*, Ar-H), 8.27 (2H, *d*, *J* = 7.9 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 35.9 (CH), 117.1, 117.8, 118.5, 123.1, 125.1, 126.4, 127.1, 128.3, 129.5, 130.1, 132.3, 136.1, 138.1, 147.4, 152.2, 159.9, 163.1 (Ar–C).

*14-(4-Hydroxyphenyl)-14*H-*dibenzo[*a,j*]xanthene* (***4j***). White crystals; Yield: 85 %; m.p.: 135–136 °C; Anal. Calcd. for $C_{27}H_{18}O_2$ (FW: 374.13): C, 86.61; H, 4.85 %. Found: C, 86.42; H, 4.98 %; FT-IR (KBr, cm^{-1}): 3441 (OH stretching), 3060 (=C–H stretching of aromatic ring), 1623 (C=C– stretching of aromatic ring), 1581, 1244 (C–O stretching), 819; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 5.54 (1H, *bs*, OH), 6.44 (1H, *s*, CH), 6.87–6.91 (4H, *m*, Ar-H), 7.12 (2H, *d*, *J* = = 7.8 Hz, Ar-H), 7.22–7.26 (4H, *m*, Ar-H), 7.39 (4H, *d*, *J* = 8.0 Hz, Ar-H), 7.52 (2H, *d*, *J* = 7.8 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 36.8 (CH), 114.9, 116.8, 118.3, 121.5, 123.8, 125.2, 126.1, 127.2, 128.9, 130.5, 132.5, 135.1, 144.2, 152.7.

*14-(3-Nitrophenyl)-14*H-*dibenzo[*a,j*]xanthene* (***4k***). Yellow crystals; yield: 90 %; m.p.: 208–210 °C; Anal. Calcd. for $C_{27}H_{17}NO_3$ (FW: 403.12): C, 80.38; H, 4.25; N, 3.47 %. Found: C, 80.61; H, 4.38; N, 3.20 %; FT-IR (KBr, cm^{-1}): 3045 (=C–H stretching of aromatic ring), 1626 (C=C– stretching of aromatic ring), 1551 (NO_2), 1344 (NO_2), 1241 (C–O stretching), 811; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 6.60 (1H, *s*, CH), 6.65 (1H, *s*, Ar-H) 7.41–7.44 (3H, *m*, Ar-H), 7.48 (2H, *d*, *J* = 7.9 Hz, Ar-H), 7.51–7.54 (4H, *m*, Ar-H), 7.64 (2H, *d*, *J* =

= 7.9 Hz, Ar-H), 7.83–7.87 (4H, *m*, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 37.3 (CH), 116.3, 116.8, 117.4, 121.1, 122.3, 125.4, 126.9, 128.2, 129.1, 134.7, 136.3, 137.1, 138.2, 146.5, 147.9, 158.3.

*14-(4-Isopropylphenyl)-14*H-*dibenzo[*a,j*]xanthene* (***4l***). Yellow crystals; yield: 85 %; m.p.: 152–154 °C; Anal. Calcd. for $C_{30}H_{24}O$ (FW: 400.18): C, 89.97; H, 6.04 %. Found: C, 89.73; H, 6.18 %; FT-IR (KBr, cm^{-1}): 3048 (=C–H stretching of aromatic ring), 1578 (C=C– stretching of aromatic ring), 1510, 1244 (C–O stretching), 806; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.22 (6H, *d*, 2×CH_3), 3.1 (1H, *m*, C**H**(CH_3)$_2$), 6.46 (1H, *s*, CH), 6.85 (2H, *d*, *J* = 8.0 Hz, Ar-H), 7.12–7.41 (4H, *m*, Ar-H), 7.48–7.53 (4H, *m*, Ar-H), 7.68–7.71 (4H, *m*, Ar-H), 7.87 (2H, *d*, *J* = 8.0 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 23.8 (2×CH_3), 33.5 (CH), 37.6 (CH), 117.6, 118.1, 122.8, 124.2, 126.4, 126.5, 126.7, 128.0, 128.8, 131.1, 131.5, 142.3, 146.6, 148.8 (Ar–C).

*4-(14*H-*dibenzo[a,j]xanthen-14-yl)benzonitrile* (***4m***). White crystals; yield: 92 %; m.p.: 209–211 °C; Anal. Calcd. for $C_{28}H_{17}NO$ (FW: 383.13): C, 87.71; H, 4.47; N, 3.65 %. Found: C, 87.58; H, 4.59; N, 3.78 %. FT-IR (KBr, cm^{-1}): 3041 (=C–H stretching of aromatic ring), 2218 (C≡N stretching), 1622, 1583 (C=C– stretching of aromatic ring), 1242 (C–O stretching), 809; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / / ppm): 6.55 (1H, *s*, CH), 7.45 (4H, *t*, Ar-H), 7.50 (2H, *d*, *J* = 8.8 Hz, Ar-H), 7.61 (4H, *q*, Ar-H), 7.85 (4H, *t*, *J* = 8.4 Hz, Ar-H), 8.28 (2H, *d*, *J* = 8.2 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 38.0 (CH), 110.3 (Ar–C), 115.9 (C≡N), 118.0, 118.6, 122.0, 124.5, 127.1, 128.8, 129.0, 129.5, 131.0, 131.1, 132.5, 148.7, 150.0 (Ar–C); MS-EI (m/z): 383.13 (M^+).

*14-(4-(Methylthio)phenyl)-14*H-*dibenzo[*a,j*]xanthene* (***4n***). Yellow crystals; yield: 85 %; m.p.: 263–265 °C; Anal. Calcd. for $C_{28}H_{20}OS$ (FW: 404.54): C, 83.14; H, 4.98 %. Found: C, 83.27; H, 4.86 %; FT-IR (KBr, cm^{-1}): 3046 (=C–H stretching of aromatic ring), 1621 (C=C– stretching of aromatic ring), 1592, 1228 (C–O stretching), 1156 (C–S), 815; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 2.81 (3H, *s*, SCH_3), 6.42 (1H, *s*, CH), 6.83 (2H, *d*, *J* = 8.1 Hz, Ar-H), 7.34–7.42 (4H, *m*, Ar-H), 7.51–7.56 (4H, *m*, Ar-H), 7.66–7.74 (4H, *m*, Ar-H), 8.25 (2H, *d*, *J* = 8.1 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 21.2 (SCH_3), 34.2 (CH), 112.1, 116.9, 117.1, 121.9, 123.9, 126.5, 127.1, 128.8, 129.1, 130.9, 131.5, 138.1, 147.5, 156.1 (Ar–C).

*4-(14*H-*Dibenzo[*a,j*]xanthen-14-yl)benzaldehyde* (***4o***). Pink crystals; yield: 90 %; m.p.: 252–254 °C; Anal. Calcd. for $C_{28}H_{18}O_2$ (FW: 386.13): C, 87.03; H, 4.69 % Found: C, 87.26; H, 4.51 %; FT-IR (KBr, cm^{-1}): 3060 (=C–H stretching of aromatic ring), 2923, 2765 (–CHO), 1691 (–C=O stretching of –CHO), 1595 (C=C– stretching of aromatic ring), 1513, 1243 (C–O stretching), 819; ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 6.58 (1H, *s*, CH), 7.18 (2H, *d*, *J* = 8.0 Hz, Ar-H), 7.43 (2H, *t*, *J* = 7.8 Hz, Ar-H), 7.50–7.68 (4H, *m*, Ar-H), 7.70–7.78 (4H, *m*, Ar-H),

7.84 (2H, *t*, *J* = 7.6 Hz, Ar-H), 8.34 (2H, *d*, *J* = 8.0 Hz, Ar-H), 9.79 (1H, *s*, CHO); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 33.5 (CH), 117.6, 118.1, 122.8, 124.2, 126.4, 126.5, 126.7, 128.7, 128.8, 131.1, 131.5, 142.3, 146.6, 148.8 (Ar-C), 192.3 (–C=O).

*3,4,6,7-Tetrahydro-3,3,6,6-tetramethyl-9-phenyl-2*H*-xanthene-1,8(*5H,9H*)-dione (**5a**)*. White solid; yield: 89 %; m.p.: 203–205 °C; Anal. Calcd. for $C_{23}H_{26}O_3$ (FW: 350.19): C, 78.83; H, 7.48 %. Found: C, 78.72; H, 7.59 %; FT-IR (KBr, cm^{-1}): 3052 (=C–H stretching of aromatic ring), 2942, 1661 (C=O), 1618 (C=C– stretching of aromatic ring), 1362, 1199 (C–O stretching); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 0.99 (6H, *s*, 2×CH_3), 1.10 (6H, *s*, 2×CH_3), 1.99––2.12 (4H, *m*, 2×CH_2), 2.46 (4H, *s*, 2×CH_2), 4.67 (1H, *s*, CH), 7.24–7.28 (5H, *m*, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 27.6 (2×CH_3), 28.2 (2×CH_3), 32.1 (2×**C**$(CH_3)_2$), 32.5 (CH), 40.8 (2×CH_2), 50.8 (2×CH_2), 110.2, 119.0, 129.2, 132.0, 149.4, 162.9 (Ar–C), 196.3 (C=O).

*9-(4-Methoxyphenyl)-3,3,6,6-tetramethyl-3,4,5,6,7,9-hexahydro-1*H*-xanthene-1,8(2*H*)-dione (**5b**)*. White solid; yield: 85 %; m.p.: 242–245 °C; Anal. Calcd. for $C_{24}H_{28}O_4$ (FW: 383.20): C, 75.76; H, 7.42 %. Found: C, 75.62; H, 7.57 %; FT-IR (KBr, cm^{-1}): 3042 (=C–H stretching of aromatic ring), 2955, 1667 (C=O), 1623 (C=C– stretching of aromatic ring), 1249 (C–O), 1198 (C–O stretching); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 0.99 (6H, *s*, 2×CH_3), 1.10 (6H, *s*, 2×CH_3), 2.14–2.25 (4H, *m*, 2×CH_2), 2.45 (4H, *s*, 2×CH_2), 3.73 (3H, *s*, OCH_3), 4.70 (1H, *s*, CH), 6.75 (2H, *d*, *J* = 8.1 Hz, Ar-H), 7.20 (2H, *d*, *J* = 8.1 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 27.3 (2×CH_3), 29.2 (2×CH_3), 30.9 (2×**C**$(CH_3)_2$), 32.3 (CH), 40.7 (2×CH_2), 50.6 (2×CH_2), 55.7 (OCH_3), 113.1, 115.5, 129.2, 136.5, 155.8, 168.1 (Ar–C), 196.6 (C=O).

*9-(4-Chlorophenyl)-3,3,6,6-tetramethyl-3,4,5,6,7,9-hexahydro-1*H*-xanthene-1,8(2*H*)-dione (**5c**)*. White solid; Yield: 88 %; m.p.: 230–233 °C; Anal. Calcd. for $C_{23}H_{25}ClO_3$ (FW: 384.15): C, 71.77; H, 6.55 %. Found: C, 71.62; H, 6.66 %; FT-IR (KBr, cm^{-1}): 3054 (=C–H stretching of aromatic ring), 2963, 1666 (C=O), 1617 (C=C– stretching of aromatic ring), 1364, 1211 (C–O stretching); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.01 (6H, *s*, 2×CH_3), 1.11 (6H, *s*, 2×CH_3), 2.14––2.25 (4H, *m*, 2×CH_2), 2.45 (4H, *s*, 2×CH_2), 4.95 (1H, *s*, CH), 7.15 (2H, *d*, *J* = = 8.1 Hz, Ar-H), 7.38 (2H, *d*, *J* = 8.1 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 27.2 (2×CH_3), 29.2 (2×CH_3), 32.1 (2×**C**$(CH_3)_2$), 32.2 (CH), 40.8 (2×CH_2), 50.9 (2×CH_2), 110.1, 119.2, 129.5, 132.1, 149.4, 162.8 (Ar–C), 196.6 (C=O).

*9-(4-Bromophenyl)-3,3,6,6-tetramethyl-3,4,5,6,7,9-hexahydro-1*H*-xanthene--1,8(2*H*)-dione (**5d**)*. White solid; yield: 88 %; m.p.: 226–229 °C; Anal. Calcd. for $C_{23}H_{25}BrO_3$ (FW: 428.10): C, 64.34; H, 5.87 %. Found: C, 64.42; H, 8.77 %; FT-IR (KBr, cm^{-1}): 3061 (=C–H stretching of aromatic ring), 2963, 1663 (C=O), 1619 (C=C– stretching of aromatic ring), 1358, 1218 (C–O stretching); ^{1}H-NMR

(400 MHz, $CDCl_3$, δ / ppm): 0.99 (6H, *s*, 2×CH_3), 1.12 (6H, *s*, 2×CH_3), 2.14–2.26 (4H, *m*, 2×CH_2), 2.46 (4H, *s*, 2×CH_2), 4.69 (1H, *s*, CH), 7.17 (2H, *d*, *J* = 8.0 Hz, Ar-H), 7.33 (2H, *d*, *J* = 8.0 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 27.1 (2×CH_3), 29.2 (2×CH_3), 32.2 (2×**C**$(CH_3)_2$), 32.3 (CH), 40.7 (2×CH_2), 50.9 (2×CH_2), 111.1, 119.1, 129.5, 132.2, 149.5, 162.6 (Ar–C), 196.4 (C=O).

*3,3,6,6-Tetramethyl-9-(4-nitrophenyl)-3,4,5,6,7,9-hexahydro-1*H*-xanthene-1,8(2*H*)-dione (**5e**)*. Yellow solid; Yield: 90 %; m.p.: 225–226 °C; Anal. Calcd. for $C_{23}H_{25}NO_5$ (FW: 395.17): C, 69.86; H, 6.37; N, 3.54 %. Found: C, 69.97; H, 6.26; N, 3.43 %; FT-IR (KBr, cm^{-1}): 3064 (=C–H stretching of aromatic ring), 1667 (C=O), 1611 (C=C– stretching of aromatic ring), 1538 (NO_2), 1344 (NO_2), 1214 (C–O stretching); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.00 (6H, *s*, 2×CH_3), 1.12 (6H, *s*, 2×CH_3), 2.12–2.24 (4H, *m*, 2×CH_2), 2.41–2.44 (4H, *m*, 2×CH_2), 4.86 (1H, *s*, CH), 7.38 (2H, *d*, *J* = 8.0 Hz, Ar-H), 7.98 (2H, *d*, *J* = 8.0 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ/ ppm): 27.2 (2×CH_3), 29.2 (2×CH_3), 32.1 (2×**C**$(CH_3)_2$), 32.2 (CH), 40.8 (2×CH_2), 50.9 (2×CH_2), 109.2, 118.8, 129.5, 132.2, 149.4, 163.1 (Ar–C), 196.9 (C=O).

*3,3,6,6-Tetramethyl-9-(*p*-tolyl)-3,4,5,6,7,9-hexahydro-1*H*-xanthene-1,8(2*H*)-dione (**5f**)*. Yellow solid; yield: 87 %; m.p.: 212–214 °C; Anal. Calcd. for $C_{24}H_{28}O_3$ (FW: 364.20): C, 79.09; H, 7.74 %. Found: C, 78.92; H, 7.89; %; FT-IR (KBr, cm^{-1}): 3022 (=C–H stretching of aromatic ring), 2956, 1666 (C=O), 1625 (C=C– stretching of aromatic ring), 1365, 1197 (C–O stretching); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.00 (6H, *s*, 2×CH_3), 1.10 (6H, *s*, 2×CH_3), 2.15––2.25 (4H, *m*, 2×CH_2), 2.28 (3H, CH_3), 2.47 (4H, *s*, 2×CH_2), 4.71 (1H, *s*, CH), 6.91 (2H, *d*, *J* = 7.9 Hz, Ar-H), 7.08 (2H, *d*, *J* = 7.9 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 22.1 (Ar–CH_3), 27.5 (2×CH_3), 28.2 (2×CH_3), 32.2 (2×**C**$(CH_3)_2$), 32.4 (CH), 40.8 (2×CH_2), 50.9 (2×CH_2), 109.1, 119.1, 129.2, 132.2, 148.1, 161.8 (Ar–C), 196.7 (C=O).

*3,3,6,6-Tetramethyl-9-(2-nitrophenyl)-3,4,5,6,7,9-hexahydro-1*H*-xanthene--1,8(2*H*)-dione (**5g**)*. Yellow solid; yield: 89 %; m.p.: 246–248 °C; Anal. Calcd. for $C_{23}H_{25}NO_5$ (FW: 395.17): C, 69.86; H, 6.37; N, 3.54 %. Found: C, 69.72; H, 6.49; N, 3.66 %; FT-IR (KBr, cm^{-1}): 3043 (=C–H stretching of aromatic ring), 1663 (C=O), 1618 (C=C– stretching of aromatic ring), 1544 (NO_2), 1347 (NO_2), 1197 (C–O stretching); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 0.95 (6H, *s*, 2×CH_3), 1.09 (6H, *s*, 2×CH_3), 2.10–2.23 (4H, *m*, 2×CH_2), 2.35–2.49 (4H, *s*, 2×CH_2), 4.86 (1H, *s*, CH), 7.11 (1H, *d*, *J* = 8.0 Hz, Ar-H), 7.17 (1H, *t*, *J* = 7.9 Hz, Ar-H), 7.38 (1H, *t*, *J* = 7.9 Hz, Ar-H), 7.79 (1H, *d*, *J* = 8.0 Hz, Ar-H); ^{13}C--NMR (100 MHz, $CDCl_3$, δ / ppm): 27.3 (2×CH_3), 28.2 (2×CH_3), 32.1 (2×**C**$(CH_3)_2$), 32.5 (CH), 40.7 (2×CH_2), 50.9 (2×CH_2), 109.1, 119.1, 122.2, 129.2, 132.2, 148.1, 158.1, 161.8 (Ar–C), 196.2 (C=O).

*9-(4-Fluorophenyl)-3,3,6,6-tetramethyl-3,4,5,6,7,9-hexahydro-1*H-*xanthene-1,8(2*H)*-dione (**5h**)*. White solid; yield: 90 %; m.p.: 225–227 °C; Anal. Calcd. for $C_{23}H_{25}FO_3$ (FW: 368.18): C, 74.98; H, 6.84 %. Found: 74.88; H, 6.73 %; FT-IR (KBr, cm^{-1}): 3068 (=C–H stretching of aromatic ring), 2953, 1666 (C=O), 1612 (C=C– stretching of aromatic ring), 1355, 1222 (C–O stretching); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.03 (6H, *s*, 2×CH_3), 1.12 (6H, *s*, 2×CH_3), 2.14––2.25 (4H, *m*, 2×CH_2), 2.45 (4H, *m*, 2×CH_2), 4.95 (1H, *s*, CH), 7.15 (2H, *d*, *J* = = 8.1 Hz, Ar-H), 7.38 (2H, *d*, *J* = 8.1Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 28.1 (2×CH_3), 29.2 (2×CH_3), 32.2 (2×**C**$(CH_3)_2$), 32.2 (CH), 40.6 (2×CH_2), 50.7 (2×CH_2), 111.5, 118.9, 123.7, 136.2, 141.1, 161.9 (Ar–C), 195.9 (C=O).

*9-(3-Chlorophenyl)-3,3,6,6-tetramethyl-3,4,5,6,7,9-hexahydro-1*H-*xanthene--1,8(2*H)*-dione (**5i**)*. White solid; Yield: 86 %; m.p.: 184–185 °C; Anal. Calcd. for $C_{23}H_{25}ClO_3$ (FW: 384.15): C, 71.77; H, 6.55 %. Found: C, 71.88; H, 6.44 %; FT-IR (KBr, cm^{-1}): 3051 (=C–H stretching of aromatic ring), 2966, 1667 (C=O), 1621 (C=C– stretching of aromatic ring), 1354, 1216 (C–O stretching); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.06 (6H, *s*, 2×CH_3), 1.17 (6H, *s*, 2×CH_3), 2.12–2.21 (4H, *m*, 2×CH_2), 2.46 (4H, *s*, 2×CH_2), 4.86 (1H, *s*, CH), 7.42–7.45 (3H, *m*, Ar-H), 7.72 (1H, *s*, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / / ppm): 27.9 (2×CH_3), 28.8 (2×CH_3), 32.2 (2×**C**$(CH_3)_2$), 32.3 (CH), 40.7 (2×CH_2), 50.9 (2×CH_2), 110.1, 119.2, 129.5, 132.1, 149.4, 152.7, 157.6, 162.8 (Ar–C), 196.6 (C=O).

*9-(4-Hydroxyphenyl)-3,3,6,6-tetramethyl-3,4,5,6,7,9-hexahydro-1*H-*xanthene-1,8(2*H)*-dione (**5j**)*. White solid; yield: 85 %; m.p.: 244–246 °C; Anal. Calcd. for $C_{24}H_{26}O_4$ (FW: 366.18): C, 75.38; H, 7.15 %. Found: C, 75.49; H, 7.07 %; FT-IR (KBr, cm^{-1}): 3448 (OH stretching), 3052 (=C–H stretching of aromatic ring), 2955, 1666 (C=O), 1611 (C=C– stretching of aromatic ring), 1208 (C–O stretching); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.01 (6H, *s*, 2×CH_3), 1.12 (6H, *s*, 2×CH_3), 2.13–2.23 (4H, *m*, 2×CH_2), 2.46 (4H, *s*, 2×CH_2), 4.70 (1H, *s*, CH), 5.48 (1H, *bs*, OH), 6.85 (2H, *d*, *J* = 8.0 Hz, Ar-H), 7.24 (2H, *d*, *J* = 8.0 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 27.7 (2×CH_3), 29.1 (2×CH_3), 30.9 (2×**C**$(CH_3)_2$), 32.2 (CH), 40.7 (2×CH_2), 50.9 (2×CH_2), 111.2, 115.4, 129.1, 133.1, 154.5, 168.2 (Ar–C), 196.9 (C=O).

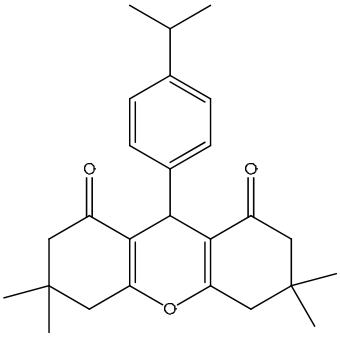
*3,3,6,6-Tetramethyl-9-(3-nitrophenyl)-3,4,5,6,7,9-hexahydro-1*H-*xanthene--1,8(2*H)*-dione (**5k**)*. Yellow solid; yield: 89 %; m.p.: 165–167 °C; Anal. Calcd. for $C_{23}H_{25}NO_5$ (FW: 384.15): C, 71.77; H, 6.55 %. Found: C, 71.88; H, 6.44 %; FT-IR (KBr, cm^{-1}): 3042 (=C–H stretching of aromatic ring), 2966, 1663 (C=O), 1623 (C=C– stretching of aromatic ring), 1541 (NO_2), 1364 (NO_2), 1218 (C–O stretching); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.01 (6H, *s*, 2×CH_3), 1.12 (6H, *s*, 2×CH_3), 2.14–2.26 (4H, *m*, 2×CH_2), 2.46 (4H, *s*, 2×CH_2), 4.81 (1H, *s*, CH), 7.55–7.57 (3H, *m*, Ar-H), 7.92 (1H, *s*, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$,

δ / ppm): 27.6 (2×CH_3), 28.9 (2×CH_3), 32.2 (2×**C**(CH_3)$_2$), 32.1 (CH), 40.8 (2×CH_2), 50.8 (2×CH_2), 110.2, 120.1, 130.4, 134.1, 150.5, 154.5, 158.1, 161.9 (Ar–C), 195.8 (C=O).

*4-(2,3,4,5,6,7,8,9-Octahydro-3,3,6,6-tetramethyl-1,8-dioxo-1*H*-xanthen-9-yl)-benzonitrile (**5m**).* Yellow solid; Yield: 90 %; m.p.: 216–217 °C; Anal. Calcd. for $C_{24}H_{25}NO_3$ (FW: 375.18): C, 76.77; H, 6.71; N, 3.73 %. Found: C, 76.92; H, 6.86; N, 3.82 %; FT-IR (KBr, cm^{-1}): 3065 (=C–H stretching of aromatic ring), 2960, 2225 (C≡N), 1663 (C=O), 1620 (C=C– stretching of aromatic ring), 1362, 1199 (C–O); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 0.99 (6H, *s*, 2×CH_3), 1.12 (6H, *s*, 2×CH_3), 2.15–2.28 (4H, *m*, 2×CH_2), 2.49 (4H, *s*, 2×CH_2), 4.77 (1H, *s*, CH), 7.42 (2H, *d*, *J* = 8.0 Hz, Ar-H), 7.53 (2H, *d*, *J* = 8.0 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 27.3 (2×CH_3), 29.2 (2×CH_3), 32.2 (2×**C**(CH_3)$_2$), 32.4 (CH), 40.8 (2×CH_2), 50.6 (2×CH_2), 110.2, 114.6 (C≡N), 119.0, 129.2, 132.0, 149.4, 162.9 (Ar–C), 196.3 (C=O); MS-EI (m/z): 375.18 (M^+).

SUPPLEMENTARY INFORMATION FOR NEW COMPOUNDS

*9-(4-Isopropylphenyl)-3,3,6,6-tetramethyl-3,4,5,6,7,9-hexahydro-1*H*-xanthene--1,8(2*H*)-dione (**5l**)*



Yellow solid; Yield: 80 %; m.p.: 203–206 °C; Anal. Calcd. for $C_{26}H_{32}O_3$ (FW: 392.24): C, 79.56; H, 8.22 %. Found: C, 79.39; H, 8.36 %; FT-IR (KBr, cm^{-1}): 3071 (=C–H stretching of aromatic ring), 2961, 1665 (C=O), 1624 (C=C– stretching of aromatic ring), 1359, 1198 (C–O); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / / ppm): 1.00 (6H, *s*, 2×CH_3), 1.10 (6H, *s*, 2×CH_3), 1.18 (6H, *d*, 2×CH_3), 2.16––2.26 (4H, *m*, 2×CH_2), 2.46 (4H, *s*, 2×CH_2), 2.78–2.81 (1H, *m*, C**H**(CH_3)$_2$), 4.73 (1H, *s*, CH), 7.05 (2H, *d*, *J* = 8.0 Hz, Ar-H), 7.18 (2H, *d*, *J* = 8.0 Hz, Ar-H); ^{13}C--NMR (100 MHz, $CDCl_3$, δ / ppm): 23.9 (2×CH_3), 27.4 (2×CH_3), 29.2 (2×CH_3), 31.3 (C**H**(CH_3)$_2$), 32.2 (2×**C**(CH_3)$_2$), 33.6 (CH), 40.8 (2×CH_2), 50.8 (2×CH_2), 115.8, 126.1, 128.1, 141.3, 146.5, 162.1 (Ar–C), 196.4 (C=O); MS-EI (*m/z*): 392.24 (M^+).

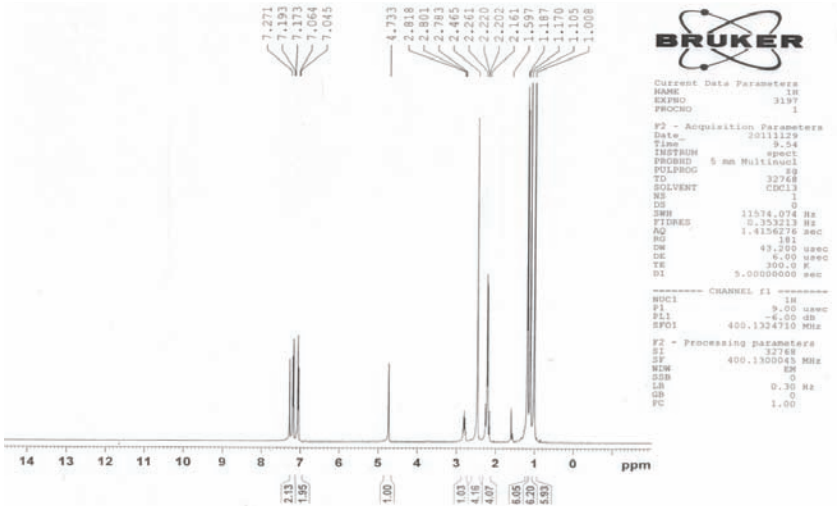


Fig. S-1. ^{1}H-NMR (400 MHz, $CDCl_3$) spectrum of **5l**.

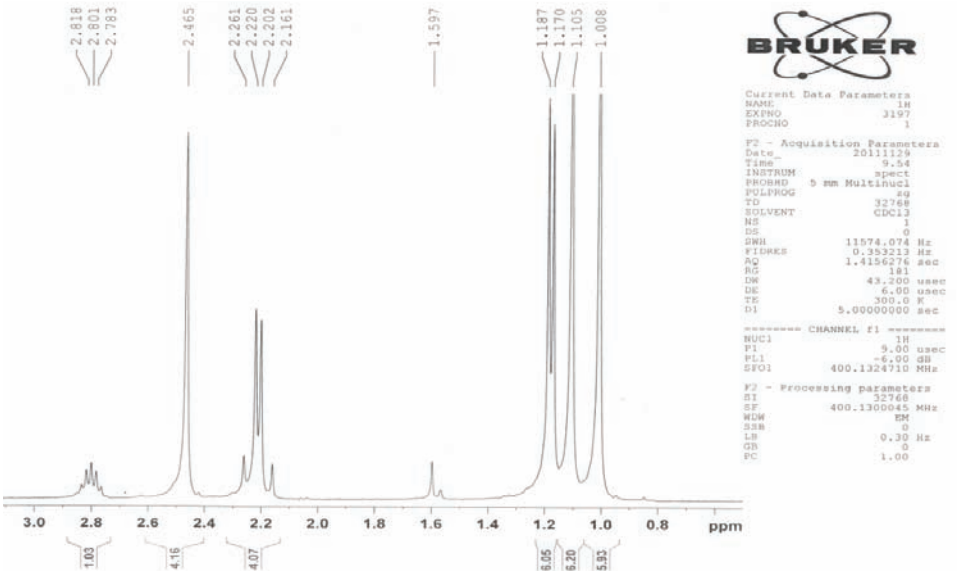


Fig. S-2. ^{1}H-NMR (400 MHz, $CDCl_3$) expanded spectrum of **5l**.

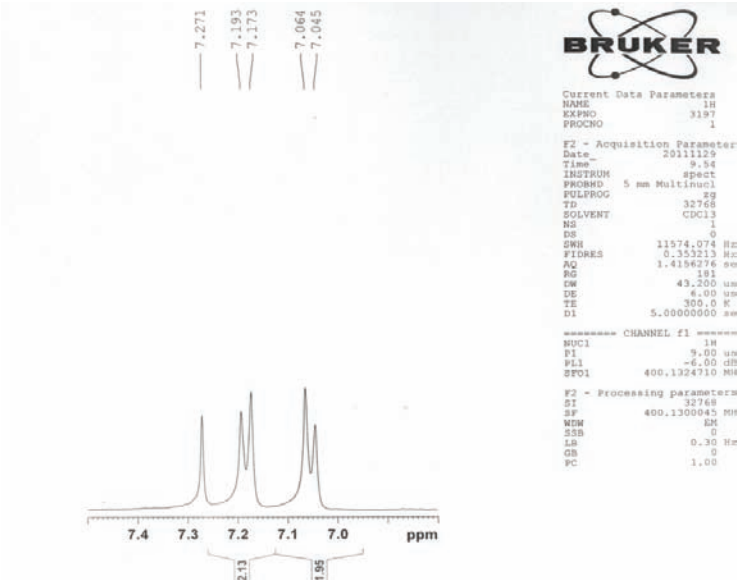


Fig. S-3. ^{1}H-NMR (400 MHz, $CDCl_3$) expanded spectrum of **5l**.

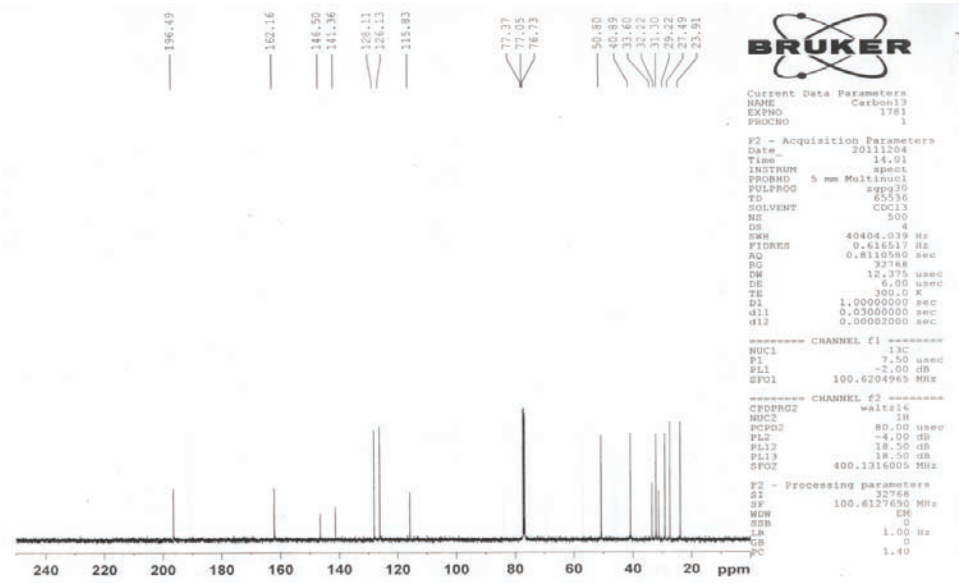
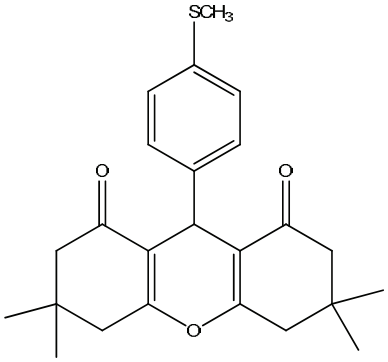


Fig. S-4. ^{13}C-NMR (100 MHz, $CDCl_3$) spectrum of **5l**.

*3,3,6,6-Tetramethyl-9-[4-(methylthio)-phenyl]-3,4,5,6,7,9-hexahydro-1*H*-xanthene-1,8(2*H*)-dione (**5n**)*



White solid; Yield: 86 %; m.p.: 256–257 °C; Anal. Calcd. for $C_{24}H_{28}O_3S$ (FW: 396.18): C, 72.69; H, 7.12 %. Found: C, 72.82; H, 6.99 %; FT-IR (KBr, cm^{-1}): 3045 (=C–H stretching of aromatic ring), 2963, 1661 (C=O), 1621 (C=C– stretching of aromatic ring), 1226 (C–O), 1152 (C–S); ^{1}H-NMR (400 MHz, $CDCl_3$, δ / ppm): 1.00 (6H, *s*, 2×CH_3), 1.10 (6H, *s*, 2×CH_3), 2.14–2.21 (4H, *m*, 2×CH_2), 2.24 (3H, *s*, SCH_3), 2.46 (4H, *s*, 2×CH_2), 4.71 (1H, *s*, CH), 7.02 (2H, *d*, *J* = 7.9 Hz, Ar-H), 7.18 (2H, *d*, *J* = 7.9 Hz, Ar-H); ^{13}C-NMR (100 MHz, $CDCl_3$, δ / ppm): 21.4 (SCH_3), 27.3 (2×CH_3), 29.2 (2×CH_3), 30.9 (2×**C**$(CH_3)_2$), 32.1 (CH), 40.8 (2×CH_2), 50.7 (2×CH_2), 113.4, 115.7, 129.3, 136.5, 157.9, 162.1 (Ar–C), 196.4 (C=O); MS-EI (*m/z*): 396.18 (M^+).

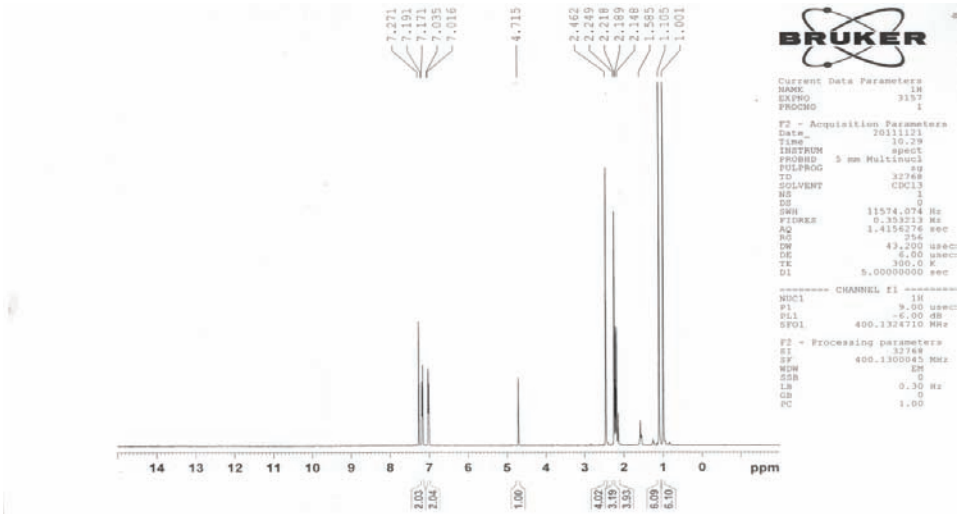


Fig. S-5. ^{1}H-NMR (400 MHz, $CDCl_3$) spectrum of **5n**.

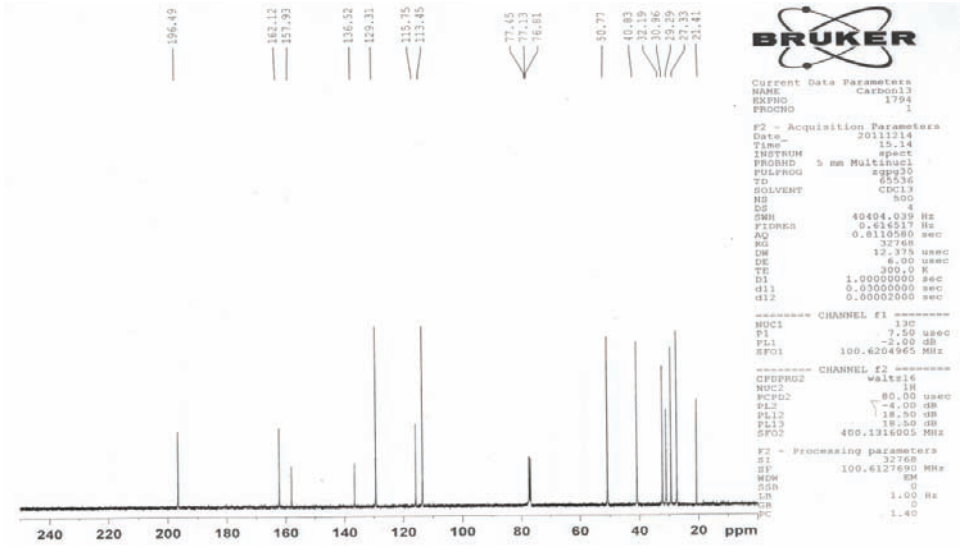
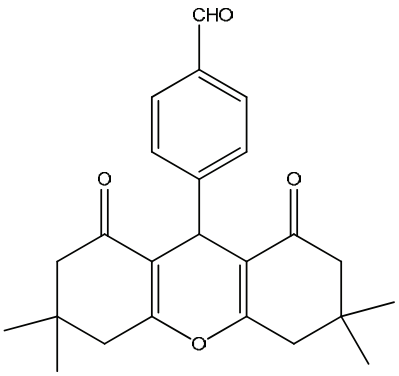


Fig. S-6. ^{13}C-NMR (100 MHz, $CDCl_3$) spectrum of **5n**.

*4-(2,3,4,5,6,7,8,9-Octahydro-3,3,6,6-tetramethyl-1,8-dioxo-1*H*-xanthen-9-yl)-benzaldehyde (**5o**)*



White solid; Yield: 89 %; m.p.: 211–213 °C; Anal. Calcd. for $C_{24}H_{26}O_4$ (FW: 378.18): C, 76.17; H, 6.92 %. Found: C, 76.05; H, 7.09 %; FT-IR (KBr, cm^{-1}): 3063 (=C–H stretching of aromatic ring), 2873 (=C–H aldehyde), 1728 (–C=O stretching of –CHO), 1663 (C=O), 1618, 1517 (C=C– stretching of aromatic ring), 1358, 1200 (C–O); ^{1}H-NMR ($CDCl_3$): 1.01 (6H, *s*, 2×CH_3), 1.10 (6H, *s*, 2×CH_3), 2.13–2.22 (4H, *m*, 2×CH_2), 2.47 (4H, *s*, 2×CH_2), 4.70 (1H, *s*, CH), 7.39 (2H, *d*, *J* = 8.0 Hz, Ar-H), 7.51 (2H, *d*, *J* = 8.0 Hz, Ar-H), 9.72 (1H, *s*, CHO); ^{13}C-NMR ($CDCl_3$): 27.3 (2×CH_3) , 29.2 (2×CH_3), 31.5 (CH), 32.2 (2×**C**$(CH_3)_2$), 40.8 (2×CH_2), 50.6 (2×CH_2), 115.1, 120.2, 130.1, 131.1, 143.2,

162.4 (Ar–C), 196.3 (–C=O), 204.8 (–C=O, aldehyde). MS-EI (*m/z*): 378.18 (M^+).

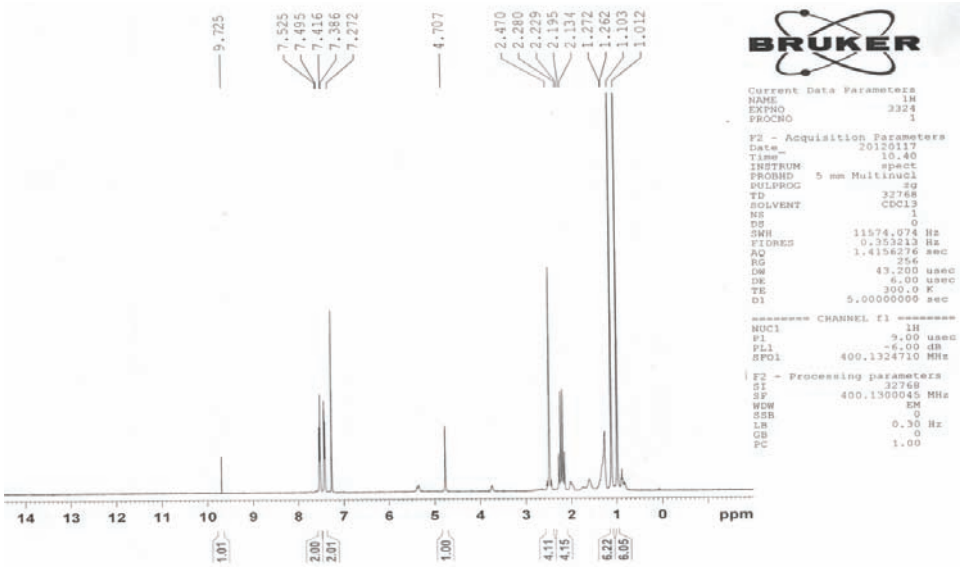


Fig. S-12. ^{1}H-NMR (400 MHz, $CDCl_3$) spectrum of **5o**.

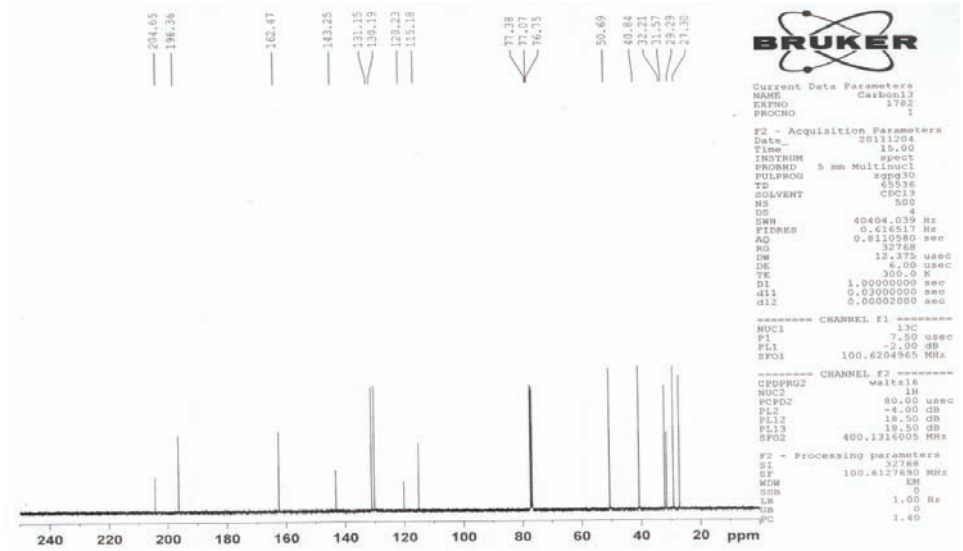


Fig. S-13. ^{13}C-NMR (100 MHz, $CDCl_3$) spectrum of **5o**.