Microwave-assisted synthesis, characterisation and mesomorphic investigations of novel disubstituted aroylhydrazones

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The present study is focused on the development of green microwave process for the synthesis of mesogenic substituted aroylhydrazones and their structure–mesophase relationship study. Two new liquid crystalline series of disubstituted aroylhydrazones with (ABB(OH)H-*) and without lateral hydroxyl group (ABBH-* (n = 6–16)) have been synthesised by microwave-assisted methods. The compounds were structurally characterised by using suitable spectroscopic techniques. The mesomorphic properties of the series of aroylhydrazones were examined using differential scanning calorimetric analysis and polarising optical microscopy. The relationship between structure and mesogenic properties was analysed by comparison of the present series and earlier reports on homostructural series of aroylhydrazones. Thus, the effects of substitution of phenyl rings with ester and ether linking group at both terminals and introduction of hydroxyl group at lateral position on stability of mesophase and their width were investigated. In general, all substituted aroylhydrazone derivatives exhibit SmC mesophase, and substitution of terminal and lateral group has considerably increased the stability and broadened the width of mesophase of the compounds.

Keywords: microwave-assisted synthesis; aroylhydrazones; structure–mesophase relationship; liquid crystals

1. Introduction

Aroylhydrazone derivatives, molecules of both biological and material science interest, are one of the earliest systems reported as components in the development of a variety of applications, such as biological, bactericidal and fungicidal drugs, and have great interest due to their analytical, industrial and pharmacological importance.[1–12] Recently, aroylhydrazones and their metal complexes have been reported as key components in antioxidant and antiproliferative activities.[13–15] First report on synthesis and mesomorphic potential of aroylhydrazone metal complexes was by McCabe et al. in 1993.[16] They reported a systematic investigation on a series of aroylhydrazinato-nickel(II) and copper(II) complexes, a novel class of metallomesogens which exhibit smectic C (SmC) and nematic (N) phases.[17] The mesomorphic nature of these complexes was found to be strongly dependent on the nature of the substituent at the azomethine moiety. In 1998, Lai et al. synthesised a series of polycatenar aroylhydrazinato-nickel(II) metallomesogens showing columnar mesophase. These derivatives showed mesophase crossover phenomenon with increasing alkoxy chain length.[18] Same year, Dalcanales and co-workers synthesised room-temperature aroylhydrazone metallomesogens by design and synthesis of various organic derivatives with different alkoxy substitution patterns.[19] Cancelli et al. adopted similar synthetic methodology and alkoxy substitution patterns as reported by Dalcanales on a novel 2,3-butane dione core. They reported columnar mesophase of aroylhydrazone metal derivatives over a good thermal range with the same structure–mesophase relationship.[20]

Mesogenic potential of aroylhydrazone derivatives was explored the first time by Yelamaggad et al. by design and synthesis of polycatenar aroylhydrazone discotics having hexagonal columnar mesophase over a wide thermal range.[21] Tschierske and Yelamaggad have reported interesting room-temperature mesophase of aroylhydrazones suitable for application purposes. The mesophase of these derivatives is due to the adaptation of unusual supramolecular helical organisation of molecules by intermolecular hydrogen bonding.[22] Similar to metal complexes, type and stability of columnar mesophase of these organic derivatives were found to be strongly dependent on the combination of specific alkoxy substitution patterns around the core group.

The effects of substituents like linking groups, lateral groups and spacers are very important in the field of structure–mesophase relationship study of liquid crystals.[23] The effect of suitable substituents on mesomorphic properties and elucidation of such relationships is important because it allows new materials to be obtained through rational design rather than serendipitous discovery.[24] The aroylhydrazone core (*–C6H5–C(=O)–NH–N=CH–C6H5–*) has
benzoyl (\(\text{C}_6\text{H}_5\text{C}=\text{O}\)) and benzylidene (\(\text{C}═\text{C}_6\text{H}_5\)) terminals for incorporation of various suitable substituent or derivatives generally used for design and synthesis of liquid crystals having predefined properties. In order to explore the possibility of calamitic mesophase in these derivatives and also to see the effect of various substituents on the aroylhydrazone core, our group has first reported a series of monoalkoxy ester-substituted aroylhydrazones showing SmC mesophase.[25] The ester group has been substituted at benzylidene moiety in these derivatives. These observations were very interesting because non-substituted derivatives were reported as non-mesogenic in nature.[17] In our another report, effect of lateral hydroxy group on mesomorphic properties of ester substituted aroylhydrazone derivatives has been observed. It was found that lateral hydroxy group has extended the mesomorphic range in such derivatives in considerable range.[26] Recently, in order to see the effect of substituents at aroylhydrazone benzoyl terminal, a very efficient microwave-assisted synthesis of a series of 4-benzylolxy-substituted aroylhydrazones has been reported.[27] The mesogenic properties of these derivatives were found to be strongly dependent on the nature of benzoyl and benzylidene substituents around the core group (Figure 1).

Microwave-assisted high-speed chemical synthesis has attracted a considerable amount of attention in the past decade. Almost all types of organic reactions have been performed using the efficiency of microwave-flash heating. This is not only due to the fact that reactions proceed significantly faster and more selectively than under conventional thermal conditions but also because of the operational simplicity, high yield of products and cleaner reactions with easier work-up.[28–32]

In continuation of our earlier work on the novel approaches on synthesis of aroylhydrazone-based liquid...
crystals and their structure–mesophase relationship study, we report herein efficient microwave-assisted synthesis of novel disubstituted aroylhydrazone derivatives. Two series of aroylhydrazone derivatives have been synthesised with alkoxy benzyl and alkoxy benzoic acid ester moiety at benzyl and benzylidene terminals, respectively, with and without lateral hydroxyl group in order to explore the simultaneous effect of substitution at both ends of aroylhydrzones and also to see the effect of lateral group on these systems.

2. Results and discussion

2.1 Synthesis

Microwave-assisted synthetic route for preparation of disubstituted aroylhydrazone-based mesogenic compounds with (ABB(OH)H-n) and without (ABBH-n) lateral hydroxyl group is outlined in Scheme 1. Initial precursors and promesogenic groups (2–9) were prepared according to our earlier reports on aroylhydrazone derivatives.[27] In the microwave-assisted method (Scheme 1), the reaction of 4-hydroxybenzoate ester (1) with appropriate alkyl bromide in the presence of cesium carbonate and N-Methyl-2-pyrrolidone (NMP) under microwave irradiation produced ethyl-4-alkoxybenzoate esters (2 and 6) in a short period of ~3–5 min. Reduction of ethyl-4-alkoxybenzoate esters by stirring in lithium aluminium hydride (LAH) at ice temperature leads to the formation of 4-alkoxybenzylalchohols (7), which on further treatment with thionyl chloride in dry dichloromethane (DCM) in the presence of one to two drops of dry DMF at ice temperature leads to the formation of 4-alkoxybenzylchlorides. Condensation of 4-alkoxybenzylchloride with ethyl 4-hydroxy ester in 1:1 molar ratio under microwave radiation, with
Cs$_2$CO$_3$ (cat.) and 3 mL of dry DMF as solvent, leads to the formation of ethyl 4-[4′-(alkoxy)benzyl]oxy benzoates (8) in high yield (82–87%). The reactions were completed in a short period of 2–3 min. 4-alkoxybenzylbenzoylhydrazines (9) were prepared by keeping properly mixed solution of ethyl 4-[4′-(alkoxy)benzyl]oxy benzoates (8) and hydrazine monohydrate in microwave oven at 80°C with mild stirring in dry ethanol as a solvent for 5–6 min of microwave irradiation. Alkoxy benzoic acid was reacted with thionyl chloride (3 equiv.) in dry DCM by refluxing for 3 h. After removal of solvent and excess thionyl chloride, 4-hydroxy benzaldehyde or 2,4-dihydroxy benzaldehyde was added and refluxed for additional 3 h to yield 4-(4′-alkoxybenzyl)oxy benzaldehydes (4) and 4-(4′-alkoxybenzyl)oxy)-2-hydroxy benzaldehydes (5). The Schiff’s bases (ABBH-n and ABB(OH)H-n) were obtained by the condensation of the hydrazine derivatives with appropriate aldehydes in 3–4 mL of dry ethanol at 80–120°C under 4–8 min of microwave irradiation with 30 s of time intervals in very good yield (75–84%).

All the intermediates and final products were thoroughly purified and fully characterised, and satisfactory analytical data were obtained. Detailed spectroscopic data for the key intermediates and final compounds are given in the supplementary information file.

### 2.2 Density functional theory (DFT) studies

The single molecular optimisation of both the compounds (ABBH-10 and ABB(OH)H-10) and their possible tautomeric forms has been done to investigate the molecular properties using the DFT method. The optimised structures of the keto and enol forms of the compounds with decyloxy chain length are shown in Figure 2. The absence of any imaginary frequency in the calculated vibrational frequencies ensures that the optimised geometry corresponds to a true energy minimum. The energies of both the keto and enol forms of the ligands were calculated. The keto form has slightly more negative energy than the enol form for both the compounds, which indicates more stability of the keto form. The room-temperature infrared (IR) and nuclear magnetic resonance (NMR) spectra of the compounds also suggest the presence of the keto form in both the solid and solution states. The optimised

Figure 2. (colour online) DFT optimised structures of keto and enol forms of (a) (ABBH-10) and (b) (ABB(OH)H-10) (method, RB3LYP, 6–311++G(d,p)).
lengths of the fully extended keto form of the compounds are found to be 51.61 and 51.33 Å, respectively. In keto form of ABBH(OH)H-10, the interaction of the O–H group with the –C=–N group through an intramolecular (O−H…N=C=O) hydrogen bond is suggested as the H…N distance (D−H…A) is found to be 1.80 Å with the phenolic-hydrogen and imine-nitrogen. The calculated dihedral angles between rings (A)–(B), (B)–(C) and (C)–(D) for ligand (ABBH-10) are 96.00°, 139.93° and 134.44°, respectively. For ligand (ABB(OH)H-10), these dihedral angles are found to be 95.99°, 139.94° and 134.99°. These values indicate twist of all phenyl rings with respect to one another, although the shape of molecules is calamitic (rod like) in nature and thus suitable for showing mesomorphic properties of calamitic molecules.

2.3 Thermal microscopy and differential scanning calorimetry: mesomorphic behaviour

The mesomorphic properties of N-[4-(4′-alkoxy)benzoyloxybenzylidene]-N′-[4″-(4″′-deceyloxy)benzoyloxybenzoyl]hydrazines (ABBH-n, n = 6–16) and N-[4-(4′-alkoxy) benzoyloxy-2-hydroxy-benzylidene]-N′-[4″′-(4″″-deceyloxy)benzoyloxybenzoyl]hydrazines (ABB (OH)H-n, n = 6–16) were investigated by differential scanning calorimetry (DSC) and polarising optical microscopy (POM). The phase transition temperature and thermodynamic data of both series of compounds are listed in Tables 1 and 2.

In the series of mesogens without lateral −OH group (ABBH-n, n = 6–16), all the compounds formed enantiotropic SmC phase. The compound (ABBH-12) with the dodecyl chain length (n = 12) at benzoyl ester moiety showed three endothermic peaks at 163.2, 209.7 and 233.9°C followed by the appearance of four exothermic peaks at 207.3, 183.1, 158.9 and 124.5°C (Figure 3a). POM observations reveal that upon heating from crystal state to 209.7°C, schlieren texture corresponding to SmC mesophase emerges (Figure 4b). Further heating of the compound leads to formation of isotropic liquid at 233.9°C. It thus becomes clear that the endothermic peak at 163.2°C in heating scan is due to crystal transformation from one state to another (Cr→Cr′ transition). In cooling cycle, the POM observations reveal the similar mesophase observed for heating cycle, i.e. schlieren SmC phase which emerges from 207.3°C and solidifies at 183.1°C (Figure 4c) and thus indicates high degree of polymorphism in the compound. The lower members of the series with hexyloxy, octyloxy and decyloxy chain length (ABBH-6, -8, -10) at benzoyl ester moiety showed similar typical schlieren texture with four brush singularities of SmC mesophase in heating cycle as well as in cooling cycles with retention of polymorphic nature.

The compound with hexadecyloxy peripheral chains (ABBH-16) showed two endothermic peaks at 207.2 and 239.1°C followed by appearance of two exothermic peaks at 211.4 and 172.7°C (Figure 3b). The two endothermic peaks observed for this compound define crystal-to-mesophase and mesophase-to-isotropic phase transitions. The exothermic peaks reveal an isotropic-to-mesophase and mesophase-to-

| Table 1. Thermal transitions and corresponding thermodynamic parameters of the compounds (ABBH-n, n = 6–16). |
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| **no.** | **Compounds** | **Heating** | **Cooling** |
| 1 | ABBH-6 | Cr 117.4 (14.3; 36.6) Cr′ 214.3 (31.5; 64.6) SmC 242.4 (6.2; 12.0) i | i 216.8 (−8.2; −16.7) SmC 187.6 (−19.2; −41.6) Cr′ 124.4 (4.6; −11.5) Cr |
| 2 | ABBH-8 | Cr 184.7 (34.2; 74.7) Cr′ 213.1 (10.5; 21.6) SmC 238.4 (8.2; 16.0) i | i 208.7 (−5.6; −11.6) SmC 183.4 (−24.6; −53.9) Cr′ 116.8 (8.3; −21.3) Cr |
| 3 | ABBH-10 | Cr 125.7 (21.3; 53.4) Cr′ 216.6 (11.4; 23.3) SmC 237.4 (9.4; 18.4) i | i 209.6 (−8.3; −17.2) SmC 185.7 (−23.5; −51.2) Cr′ 163.7 (14.6; −33.4) Cr′ 110.6 (8.7; 22.6) Cr |
| 4 | ABBH-12 | Cr 163.2 (8.3; 19.0) Cr′ 209.7 (55.4; 114.7) SmC 233.9 (15.8; 31.1) i | i 207.3 (−3.3; −6.8) SmC 183.1 (−11.8; −25.8) Cr′ 158.9 (1.9; −4.4) Cr′ 124.5 (2.8; 7.0) Cr |
| 5 | ABBH-14 | Cr 202.8 (23.2; 48.7) SmC 235.4 (4.8; 9.4) i | i 215.3 (−3.9; −7.9) SmC 178.4 (−8.2; −18.1) Cr |
| 6 | ABBH-16 | Cr 207.2 (45.4; 94.5) SmC 239.1 (6.9; 13.4) i | i 211.4 (−4.3; −8.8) SmC 172.7 (−7.5; −16.8) Cr |

Note: Cr = crystalline state, SmC = smectic C mesophase, i = isotropic liquid.
A salient enantiotropic smectic C mesophase with schlieren texture is revealed for this compound by POM. The lower member of this series with tetradecyloxy chain length (ABBH-14) showed similar behaviour of phase transitions with no observations of Cr–Cr′ transitions.

The comparative phase diagram of the compounds (ABBH-n) (Figure 6a) indicates that a very high degree of supercooling effect (~20–30°C) is found for all the compounds. It is also found that both melting and clearing temperatures are inconsistent with the length of the side chains, and a random order of mesophase transitions is found for this series of compounds. The melting and clearing temperatures all ranged between 202.8 and 216.6°C, and 233.9 and 242.4°C, respectively, in the heating cycle.

In the series of mesogens with lateral –OH group (ABB(OH)H-n, n = 6–16), all the compounds formed enantiotropic SmC phase similar to that of earlier compounds. The compound (ABB(OH)H-8) showed three endothermic peaks at 162.5, 178.0 and 239.7°C followed by the appearance of two exothermic peaks at 214.8 and 142.2°C (Figure 5a). POM observations reveal that upon heating from crystal state to 178.0°C, schlieren texture corresponding to SmC mesophase emerges (Figure 4d). Further heating of the
compound leads to the formation of isotropic liquid at 239.7°C. In the cooling cycle, a similar texture is obtained with a large degree of supercooling effect similar to that of compounds ABBH-$n$. The next higher member of the series with decyloxy chain length (ABB(OH)H-10) showed similar transition and mesophase behaviour to that of its lower member, although a significant decrease in melting ($T_m$) and isotropisation temperature ($T_i$) is found for this compound (ABB(OH)H-10) and also higher members of the series (ABB(OH)H-12, -14, -16) as shown in Figure 5b. Decrease in melting ($T_m$) and

Figure 4. (colour online) Microphotographs of the compounds ABBH-$n$ and ABB(OH)H-$n$ in SmC phase. (a) Schlieren texture of ABBH-6 at 180.1°C in heating cycle, (b) schlieren texture of ABBH-12 at 221.4°C in heating cycle, (c) schlieren texture of ABBH-12 at 196.5°C in cooling cycle, (d) schlieren texture of ABB(OH)H-8 at 216.5°C in heating cycle and (e) schlieren texture of ABB(OH)H-16 at 177.3°C in cooling cycle.

Figure 5. DSC thermograms of the compounds ABB(OH)H-$n$. (a) $n = 8$, (b) $n = 10$ and (c) $n = 16$. 
isotropisation temperature ($T_i$) of the compounds indicates the plasticisation effect after chain length ($n = 8$) and reduction in intermolecular phenomenon in the molecular domain. This affects the interaction between the molecules and consequently reduced packing behaviour and results in lowering in melting ($T_m$) and isotropisation temperature ($T_i$) of the compounds. The compound with highest alkoxy chain length (ABB(OH)H-16) showed three exo- and endothermic peaks in DSC scans indicating two Cr–Cr' transitions (polymorphism) in such compound with very good mesophase thermal range (Figures 4e and 5c). This behaviour is similar to that of lower member of the compound (ABB(OH)H-14) with teradeoxy chain length. Therefore, it is concluded that the degree of polymorphism increases in higher member of the series (ABB(OH)H-14, -16), which is in contrast to the compounds of series (ABBH-n) (Table 2).

The comparison of mesogenic properties of the compounds of series ABBH-n with ester and benzyl ether moiety present together and the compounds of series ABB(OH)H-n with ester and benzyl ether moiety and an additional lateral hydroxyl group to that of series of the compounds without ester and lateral substitution as reported earlier [27] can lead us to reveal the effects of ester as well as lateral hydroxyl group substitution on mesomorphic properties of aroylhydrazones. One similarity in the mesomorphic behaviour of the all the series of above compounds is exhibition of SmC mesophase. The effect of ester and lateral –OH group substitution can be clearly observed on the liquid crystalline property of the compounds. The lowest and highest transition temperatures of SmC mesophase of (ABB(OH)H-n) series with lateral –OH are ranged between ~60°C and 80°C, whereas those of the series of compounds (ABBH-n) without lateral group are ranged between ~20°C and 30°C. So lateral –OH group increases the mesomorphic range of the compounds by approximately two to three times. Conclusively, incorporation of ester and lateral hydroxyl group has influenced drastically the mesomorphic stability of such compounds and is found to be approximately four to nine times more in comparison to simple p-benzyloxy benzoylhydrazones (Figure 6).

3. Summary
In this article, we have reported two new series of calamitic disubstituted aroylhydrazones with and without a lateral hydroxyl group that exhibit smectic C mesophase. A new, efficient and environment-friendly approach to the synthesis of aroylhydrazone-based liquid crystals under microwave irradiation has been developed. Further, substitution of terminal and lateral substituents has a considerable effect on the mesomorphic stability of the aroylhydrazone-based compounds. The substituted aroylhydrazone with ester and ether linking group and lateral hydroxyl group presented in this report have effectively increased the
4. Experimental

4.1 Materials

Ethyl 4-hydroxybenzoate, 4-hydroxy benzaldehyde, 2,4-dihydroxy benzaldehyde, hydrazine monohydrate, LiAlH₄, thionyl chloride, potassium iodide, anhydrous Cs₂CO₃ and bromoalkanes purchased from Aldrich Chemicals (Sigma-Aldrich Corp., St. Louis, MO, USA) were used as received. All other solvents and reagents were purchased from Merck (Darmstadt, Germany). The solvents were dried using standard methods [33] when required.

4.2 Techniques and computational details

Elemental analyses were performed on a CE-440 Exeter Analytical CHN analyser. IR spectra (4000–100 cm⁻¹) were recorded on a Varian 3100 FT-IR Excalibur series spectrophotometer. ¹H and ¹³C NMR spectra were obtained on a JEOL FT-NMR AL 300 MHz spectrometer using tetramethylsilane as the internal standard. Electronic spectra were recorded on an (UV)-1700 Pharma Spec. Shimadzu UV–Vis spectrophotometer. DSC thermograms were recorded with a Mettler Toledo TC 15 TA differential scanning calorimeter at the rate of 5.0 K min⁻¹ under a nitrogen atmosphere using spec pure grade indium as a standard, by taking samples in close-lid aluminium pans. The transition temperatures from the DSC thermograms have been determined to an accuracy of ±0.1 K. The mesophase type was identified by visual comparison with known phase standards using an HT 30.01 NTT 268 Lomo polarising optical microscope fitted with a hot stage, with a temperature-controlling accuracy of 0.1 K.

Microwave reactions were performed in a CEM Discover BenchMate single-mode microwave reactor with a new sealed pressure regulation 10-mL pressurised vial with ‘snap-on’ cap and Teflon-coated magnetic stir bar. The standard temperature control system for the Discover System consists of a non-contact calibrated IR sensor, which monitors and controls the temperature conditions of the reaction vessel located in the instrument cavity.

The GAUSSIAN-09 program [34] package was employed to carry out DFT [35–40] calculations at the Becke’s three-parameter functional and Lee, Yang and Parr correlation functional (B3LYP) level [40–43] of calculation, and the 6–311++G(d,p) basis set [44–48] was used for ground state geometry optimisation and frequency calculation of the ligands with decyloxy peripheral chains (ABBH-n, ABB(OH)H-n, n = 10). The internal coordinates of the system, which are used as input for GAUSSIAN-09 program, were generated by the GAUSS VIEW 4.1 program.[49]

4.3 Synthesis of N′-[4-(4′-alkoxy) benzoyloxybenzylidene]-N′′-[4′′-(decyloxy) benzoyloxy benzoyl]hydrazines, ABBH-n

The ligands ABBH-n were prepared by irradiating absolute ethanol solution containing 4-[4′(decyloxy) benzoyloxy]benzohydrazide (1.0 equiv.) and 4-(4′-alkoxy benzoyloxy) benzaldehyde (1.0 equiv.) and few drops of acetic acid for ~5 min (30 s × 10). The solution was left overnight in the vial closed with rubber septum. The microcrystalline colourless solid was filtered off by suction, thoroughly washed with cold ethanol and recrystallised from ethanol and dried at room temperature.
4.4 Synthesis of N-[4-(4′-alkoxy)benzoylxy-2-hydroxy-benzylidene]-N′-[4′-(4′′-decyloxy)benzoyloxybenzyl]hydrazines (ABB(OH)H-n)

The ligands ABB(OH)H-n were prepared by condensation of 4-(4′-alkoxybenzoylxy)-2-hydroxy-benzaldehyde (1.0 equiv.) and 4-[4′-(decyloxy)benzoyloxy]-benzoylhydrazide (1.0 equiv.) using the similar procedure described for ABBH-n.

4.4.1 N-[4-(4′-decyloxy)benzoylxy-2-hydroxy-benzylidene]-N′-[4′-(4′′-decyloxy)benzoyloxybenzyl]hydrazine (ABB(OH)H-10)

Yield: 84 %. IR (KBr, cm⁻¹): 3460 ν s (O–H), 3304 ν s (N–H), 2923, 2858 (aliphatic C–H), 1731 (ester, C=O), 1662 (amide-I, C=O), 1580 (amide-II), 1608, 1514, 1492 (Ph), 1537, 1280, 1256 (OPh): 1H NMR (300 MHz, CDCl₃, 25°C): δ 11.09 (s, 1H, –OH), 8.98 (s, 1H, –NH), 8.40 (s, 1H, –CH=N), 8.10 (d, J(H,H)=8.1 Hz, 2H, –C₆H₄), 7.80 (d, J(H,H)=8.4 Hz, 2H, –C₆H₄), 7.32 (d, J(H,H)=8.1 Hz, 2H, –C₆H₄), 6.98 (d, J(H,H)=8.1 Hz, 2H, –C₆H₄), 6.62 (m, 4H, –C₆H₄), 6.67 (d, J(H,H)=8.1 Hz, 2H, –C₆H₄), 6.79 (s, 1H, C₆H₃), 5.03 (s, 2H, Ar–CH₃,–O–Ar), 4.03 (t, 2H, –OCH₂), 3.96 (t, 2H, –OCH₂), 1.77 (m, 4H, –OCH₂CH₂), 1.52–1.28 (m, 28H, –CH₂), 0.87 (t, 6H, –CH₃); 13C NMR (75 MHz, CDCl₃, 25°C): δ 166.55, 164.10, 163.74, 163.28, 162.68, 155.42, 152.29, 133.55, 132.38, 131.06, 128.37, 121.15, 121.07, 116.86, 114.37, 113.42, 110.75, 68.37, 52.19, 31.88, 29.54, 29.34, 29.08, 25.96, 22.67, 14.10; Elemental analyses: calculated for C₄₈H₆₂N₂O₁₂ (%), C, 74.01; H, 8.02; N, 3.60; Found, C, 73.82; H, 7.91; N, 3.40.

All the other members of the series were prepared in a similar manner. The yield, IR, NMR and elemental data for the compounds are summarised in the supplementary information file.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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Supplemental Data

Supplemental data for this article can be accessed here.

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