Synthesis and Nematicidal Bioevaluation of Substituted 2H-1-Benzopyran-2-ones and their Carbamate Derivatives Against Root-Knot Nematode (*Meloidogyne javanica*)

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Synthesis of 7-hydroxy-4,5-methyl/7-hydroxy-4-methyl/7,8-dihydroxy-4-methyl/6-chloro-7-hydroxy-4-methyl-2H-1-benzopyran-2-ones (VI-IX) have been carried out by Pechmann reaction. The condensation of synthesized 2H-1-benzopyran-2-ones (VI-X) with phenyl isocyanate (XI) gave 4,5-methyl/4-methyl/4-methyl/6-chloro-4-methyl-2-oxo-2H-benzopyran-7-yl/7,8-diyl/4-yl/phenyl carbamates (XII-XVI). The synthesized compounds were characterized on the basis of analytical and spectral data. All the compounds were evaluated for their nematicidal activity *in vitro* against second stage juveniles (J_2) of root-knot nematode (*Meloidogyne javanica*).

Keywords: Substituted-2H-1-benzopyran-2-one, Carbamate, Nematicidal activity, Root-knot nematode, *Meloidogyne javanica*.

INTRODUCTION

2H-1-benzopyran-2-ones are versatile intermediate for the synthesis of various organic compounds. They also exhibit diverse biological activities¹⁻³. In present study, we have utilized hydroxyl substituted 2H-1-benzopyran-2-ones as starting material for the synthesis of carbamates. Carbamates are derivatives of carbamic acid, HOC(O)NH_2 and are widely used as insecticides. These were originally extracted from the calabar bean, which grows in west Africa. Physostigmine, a methyl carbamate ester was isolated and characterized from the extract of this bean⁴. The synthesis of carbamates are of particular interest due to their usefulness as agrochemicals like herbicides, fungicides and antimicrobial agents⁵. Carbamates are also used in pharmaceuticals industry as drug intermediates⁶. They are generally not persistent in the environment for long time.

Among different pests, the plant parasitic nematodes cause significant losses in major crops like oil seed, cereals, pulses, sugarcane, fruit crops and vegetables. In addition to directly causing crop losses, nematodes can vector many plant viruses. Many species of nematodes exist and they attack an enormous variety of plant species. The *Meloidogyne* spp. of root-knot nematodes are economically important, since these are polyphagous, vastly distributed and sedentary endoparasites in nature. Among *Meloidogyne* sp., *Meloidogyne javanica* is most common species of root knot nematodes. Carbamates are commonly used due to their chemical stability towards acids, bases and hydrogenation⁷. Carbamates inhibit acetyl cholinesterase (AChE) in the nervous system and thereby, disrupt nervous transmission at that location⁸.

The development of synthetic pesticides revolutionized pest management in agriculture. In view of the diverse biological activities shown by carbamates, their importance as agrochemicals and as a part of our research⁹⁻¹¹, the synthesis and nematicidal activity of substituted 2-oxo-2H-1-benzopyran-7-yl/7,8-diyl/4-yl phenyl carbamates have been carried out and results are reported in this paper.

EXPERIMENTAL

The melting points were determined in open capillaries on a Ganson electrical melting point apparatus. Homogeneity of the compounds was routinely checked on silica gel-G TLC plates using ethyl acetate:hexane (3:7) as irrigant. IR spectra were recorded on Perkin Elmer FTIR spectrophotometer. The NMR spectra were recorded on Bruker AC-400-F (400 MHz) NMR spectrophotometer in CDCl_3 or $\text{DMSO}-d_6$ using tetramethylsilane (TMS) as internal reference. The chemical shift values are expressed in δ (ppm) units while J values in Hz and are compatible with the assigned structures. The elemental analyses were within ± 0.4 % of that of evaluated values. Only those spectral data have been mentioned which have a direct bearing on the assignment of the structures and are discussed here.

7-Hydroxy-4,5-dimethyl-2H-1-benzopyran-2-one (VI): A mixture of orcinol (I, 2.48 g, 20 mmol) and benzyl acetoacetate (V, 3.84 g, 20 mmol), was added portion wise to H_2SO_4 (30 mL, 73 %) with constant stirring in cold condition. The

reaction mixture was left overnight and mixed with crushed ice to get a solid product, filtered, dried and crystallized from methanol to get **VI**, yield 80 %, m.p. 253-255 °C (lit. 251-253 °C)¹². IR (nujol, $\nu_{\max}$, cm^{-1}): 3390 (OH), 1685 (C=O), 1365 (C-CH₃). ¹H NMR (400 MHz, DMSO-*d*₆): 2.28 (s, 3H, C₄-CH₃); 2.56 (s, 3H, C₅-CH₃); 5.91 (s, 1H, C₃-H); 6.51 (s, 1H, C₈-H); 6.56 (s, 1H, C₆-H). Analysis found: C, 69.61; H, 5.29 %; C₁₁H₁₀O₃ Required: C, 69.46; H, 5.30 %.

Compounds **VII-IX** were prepared similarly from **II-IV** and **V** respectively.

7-Hydroxy-4-methyl-2H-1-benzopyran-2-one (VII): Yield 78 %, m.p. 183 °C (lit. 185-186 °C)¹³. IR (nujol, $\nu_{\max}$, cm^{-1}): 3400 (OH), 1671 (C=O), 1390 (C-CH₃). ¹H NMR (400 MHz, DMSO-*d*₆): 2.38 (s, 3H, C₄-CH₃); 6.03 (s, 1H, C₃-H); 6.71 (s, 1H, C₈-H); 6.80 (d, *J* = 7.5 Hz, 1H, C₆-H); 7.48 (d, *J* = 7.5 Hz, 1H, C₅-H). Analysis found: C, 68.12; H, 4.55 %. C₁₀H₈O₃ Required: C, 68.18; H, 4.58 %.

7,8-Dihydroxy-4-methyl-2H-1-benzopyran-2-one (VIII): Yield 78 %, m.p. 239 °C (lit. 235-237 °C)¹⁴. IR (nujol, $\nu_{\max}$, cm^{-1}): 3231 (OH), 1651 (C=O), 1371 (C-CH₃). ¹H NMR (400 MHz, DMSO-*d*₆): 2.30 (s, 3H, C₄-CH₃); 6.05 (s, 1H, C₃-H); 6.95-7.75 (m, 4H, Ar-H). Analysis found: C, 62.23; H, 4.22 %. C₁₀H₈O₄ Required: C, 62.50; H, 4.20 %.

6-Chloro-7-hydroxy-4-methyl-2H-1-benzopyran-2-one (IX): Yield 82 %, m.p. 268-270 °C. IR (nujol, $\nu_{\max}$, cm^{-1}): 3370 (OH), 1678 (C=O), 844 (C-Cl), 1605 (C=C), 1390 (C-CH₃). ¹H NMR (400 MHz, DMSO-*d*₆): 2.37 (s, 1H, C₄-CH₃); 6.08 (s, 1H, C₃-H); 6.89 (s, 1H, C₅-H); 7.55 (s, 1H, C₈-H). Analysis found: C, 57.36; H, 3.37. C₁₀H₇O₃Cl Required: C, 57.03; H, 3.35.

4-Hydroxy 2H-1-benzopyran-2-one (X)¹⁵: A mixture of malonic acid monophenyl ester (180 mg, 1 mmol) and Eaton's reagent (3 mL) was stirred at 70 °C for 1 h and then water was added to this mixture while stirring vigorously. Completion of the reaction was monitored by TLC. The precipitate was filtered, washed with water and dried to give a solid. It was recrystallized from ethanol to afford **X**. Yield 80 %, m.p. 209-210 °C. IR (nujol, $\nu_{\max}$, cm^{-1}): 3380 (OH), 1680 (C=O). ¹H NMR (400 MHz, DMSO-*d*₆): 5.56 (s, 1H, C₃-H); 7.19-7.25 (m, 2H); 7.48-7.53 (m, 1H); 7.72 (d, 1H, *J* = 8.0 Hz); 11.95 (s, 1H, OH). Analysis found: C, 66.17; H, 3.76 %. C₉H₆O₃ Required: C, 66.67; H, 3.73 %.

4,5-Dimethyl-2-oxo-2H-1-benzopyran-7-yl phenylcarbamate (XII): A mixture of 7-hydroxy-4,5-dimethyl-2H-1-benzopyran-2-one (**VI**, 3.80 g, 20 mmol) and phenylisocyanate (**XI**, 2.38 g, 20 mmol) in dry benzene (20 mL) was refluxed for 12 h on steam bath. Completion of the reaction was monitored by TLC. It was then concentrated under vacuum, to give solid residue which was crystallized from methanol to afford **XII**. Yield: 78 %, m.p. 218-220 °C. IR (nujol, $\nu_{\max}$, cm^{-1}): 3370 (NH), 1690 (NH-CO), 1680 (C=O). ¹H NMR (400 MHz, DMSO-*d*₆): 2.29 (s, 3H, C₄-CH₃); 2.55 (s, 3H, C₅-CH₃); 5.93 (s, 1H, C₃-H); 6.53 (s, 1H, C₆-H); 6.92 (s, 1H, C₈-H); 7.22-7.46 (m, 5H, Ar-H); 8.50 (s, 1H, NH). Analysis found: C, 69.54; H, 4.82; N, 4.51 %. C₁₈H₁₅NO₄ Required: C, 69.89; H, 4.89; N, 4.53 %.

Other compounds **XIII-XVI** were prepared similarly from **VII-X** and **XI** respectively.

4-Methyl-2-oxo-2H-1-benzopyran-7-yl phenylcarbamate (XIII): Yield 76 %, m.p. 138-140 °C. IR (nujol, $\nu_{\max}$, cm^{-1}): 3400 (NH), 1695 (NH-CO), 1680 (C=O). ¹H NMR (400 MHz, DMSO-*d*₆): 2.45 (s, 3H, C₄-CH₃); 5.94 (s, 1H, C₃-H); 6.89 (s, 1H, C₈-H); 6.97 (s, 1H, C₆-H); 6.92-7.48 (m, 5H, Ar-H); 8.49 (s, 1H, NH). Analysis found: C, 69.34; H, 4.40; N, 4.69 %; C₁₇H₁₃NO₄ Required: C, 69.15; H, 4.44; N, 4.74 %.

4-Methyl-2-oxo-2H-1-benzopyran-7,8-diyl bis(phenylcarbamate) (XIV): Yield 74 %, m.p. 226-227 °C. IR (nujol, $\nu_{\max}$, cm^{-1}): 3392 (NH), 1695 (NH-CO), 1670 (C=O). ¹H NMR (400 MHz, DMSO-*d*₆): 2.39 (s, 3H, C₄-CH₃); 5.88 (s, 1H, C₃-H); 6.93 (s, 1H, C₅-H); 7.09-7.51 (m, 10H, Ar-H); 7.98 (s, 1H, NH). Analysis found: C, 66.47; H, 4.20; N, 6.36 %. C₂₄H₁₈N₂O₆ Required: C, 66.97; H, 4.22; N, 6.51 %.

6-Chloro-4-methyl-2-oxo-2H-1-benzopyran-7-yl phenylcarbamate (XV): Yield 78 %, m.p. 268-269 °C. IR (nujol, $\nu_{\max}$, cm^{-1}): 3400 (NH), 1700 (NH-CO), 1668 (C=O), 754 (C-Cl). ¹H NMR (400 MHz, DMSO-*d*₆): 2.36 (s, 3H, C₄-CH₃); 6.09 (s, 1H, C₃-H); 6.87 (s, 1H, C₈-H); 6.93 (s, 1H, C₅-H); 6.96-7.59 (m, 5H, Ar-H); 8.55 (s, 1H, NH). Analysis found: C, 61.71; H, 3.66; N, 4.27 %. C₁₇H₁₂NO₄Cl Required: C, 61.92; H, 3.67; N, 4.25 %.

2-Oxo-2H-1-benzopyran-4-yl phenylcarbamate (XVI): Yield 80 %, m.p. 187-188 °C. IR (nujol, $\nu_{\max}$, cm^{-1}): 3380 (NH), 1710 (NH-CO), 1695 (C=O). Analysis found: C, 68.28; H, 3.69; N, 4.94 %. C₁₆H₁₁NO₄ Required: C, 68.32; H, 3.94; N, 4.98 %.

Nematicidal bioevaluation: The plant parasitic nematode *Meloidogyne javanica* was used as test organism. Stock solutions of 2000 mg L⁻¹ of all the compounds were prepared by dispersing these in acetone. Nematicidal activity was evaluated against second stage juveniles (J₂) of *M. javanica*. A suspension of juveniles (1 mL) was poured into 5 cm Petri-dishes. Measured quantities of stock solution were added to these Petri-dishes to make final concentrations of 1000, 500, 250 and 125 ppm. Acetone and water were used as control. Each treatment was replicated three times. These Petri-dishes were kept in BOD incubator at 28 ± 1 °C. Observations were recorded after 24 h and 48 h by counting live (active) and dead (inactive) J₂s under a stereoscopic binocular microscope and the per cent mortality was counted¹⁶. The revival of immobilized nematodes was examined by randomly transferring ten J₂s to water for 24 h. None of those immobilized J₂s revived. The experimental data was statistically analyzed using two factorial completely randomized design; the compounds and the concentrations constituting the two factors.

RESULTS AND DISCUSSION

Pechmann condensation of orcinol (**I**), resorcinol (**II**), pyrogallol (**III**) and 4-chlororesorcinol (**IV**) with benzyl acetoacetate (**V**), gave their corresponding 7-hydroxy-4, 5-dimethyl-2H-1-benzopyran-2-one (**VI**), 7-hydroxy-4-methyl-2H-1-benzopyran-2-one (**VII**), 7, 8-dihydroxy-4-methyl-2H-1-benzopyran-2-one (**VIII**) and 6-chloro-7-hydroxy-4-methyl-2H-1-benzopyran-2-one (**IX**) (**Scheme-I**).

Their ¹H NMR spectra were in accordance with the proposed structures. In the spectrum (400 MHz) of 7-hydroxy-4-methyl-2H-1-benzopyran-2-one (**VII**), the diagnostic proton

Reagents and reaction conditions

H_2SO_4 (73%), Stirring

Compound no.	R^1	R^2	R^3
I, VI	H	H	CH_3
II, VII	H	H	H
III, VIII	OH	H	H
IV, IX	H	Cl	H

Scheme I: Synthesis of various substituted 2*H*-1-benzopyran-2-one

at position 3 appeared as singlet at 6.03 δ . A clean singlet of $\text{C}_4\text{-CH}_3$ was appeared at 2.38 δ followed by another singlet at 6.71 δ of C_8 position proton. Two aromatic protons $\text{C}_5\text{-H}$ and $\text{C}_6\text{-H}$ appeared downfield as doublets ($J = 7.0$ Hz) integrating for one proton each at 6.80 δ and 7.48 δ respectively. Similarly ^1H NMR spectra of 7-hydroxy-4,5-dimethoxy-2*H*-1-benzopyran-2-one (**VI**) showed two singlets at 2.28 δ and 2.56 δ for $\text{C}_4\text{-CH}_3$ and $\text{C}_5\text{-CH}_3$ moiety along with other protons at their usual positions. The presence of a peak about 3400 cm^{-1} for hydroxyl group and about 1700 cm^{-1} for coumarin moiety in its IR spectrum further corroborated the assigned structure.

In view of great potential in 2*H*-1-benzopyran-2-one and their corresponding carbamates, the condensation of **VI-X** with phenylisocyanate (**XI**) by refluxing in dry benzene afforded their corresponding 4,5-dimethyl-2-oxo-2*H*-1-benzopyran-7-yl phenyl carbamate (**XII**), 4-methyl-2-oxo-2*H*-1-benzopyran-7-yl phenyl carbamate (**XIII**), 4-methyl-2-oxo-2*H*-1-benzopyran-7,8-diyl *bis*-(phenylcarbamate) (**XIV**), 6-chloro-4-methyl-2-oxo-2*H*-1-benzopyran-7-yl phenyl carbamate (**XV**) and 2-oxo-2*H*-1-benzopyran-4-yl phenyl carbamate (**XVI**) in good yields (**Scheme-II**).

In ^1H NMR spectrum of 4,5-dimethyl-2-oxo-2*H*-1-benzopyran-7-yl phenyl carbamate (**XII**), the two protons at position 6 and 8 appeared at 6.53 δ and 6.92 δ as singlets respectively. The upfield shift of $\text{C}_6\text{-H}$ proton ortho to carbamoyloxy moiety

Reagents and reaction conditions:

Benzene, Δ

Compound no.	R^1	R^2	R^3
VI, XII	H	H	CH_3
VII, XIII	H	H	H
VIII, XIV	OH	H	H
IX, XV	H	Cl	H
XIV	COONH-Ph	H	H

Reagents and reaction conditions

Benzene, Δ

Scheme-II: Synthesis of various substituted 2-oxo-2*H*-1-benzopyran-7-yl/7,8-diyl/4-yl phenyl carbamate

appeared to be a consequence of the shielding effect of carbamate functionality. Two singlets at 2.29 δ and 2.55 δ integrating for three protons each were assigned to methyl moiety at positions 4 and 5 respectively. The remaining aromatic protons were appeared as multiplet at 7.22 δ - 7.46 δ . The broad singlet at 8.50 δ was assigned to NH proton of carbamoyloxy moiety. The formation of carbamates of the above compounds follow from the mode of synthesis and were supported by the appearance of bands around 3340 cm^{-1} for NH functionality and around 1710 cm^{-1} and 1690 cm^{-1} for 2-pyrone and carbamoyloxy (C=O) functionality respectively. Thus, the structures of all these compounds were fully supported by their NMR and IR spectra.

Nematicidal bioevaluation: Nematicidal activity of all the synthesized compounds was evaluated against second stage juveniles (J_2) of *M. javanica* at four different concentrations viz. 1000, 500, 250 and 125 ppm. Acetone with water was used as control. No nematode mortality was recorded in these controls and is therefore not included in the Table-1.

The data of nematicidal activity of various substituted 2*H*-1-benzopyran-2-one against *M. javanica* J_2 s after 24 h is given in Table-1. The interaction of compounds and concentrations was statistically significant. Compound 6-chloro-7-hydroxy-4-methyl-2*H*-1-benzopyran-2-one (**IX**) showed 100 % mortality at all tested concentrations and proved to be highly nematotoxic. The presence of chloro group in 2*H*-1-benzopyran-2-one is highly effective and resulted in complete mortality of nematodes. Further evaluation of this compound was also done at 120, 60, 30, 15 and 7.5 ppm. It has shown mortality at lowest concentration also *i.e.*, 7.5 ppm.

So due to its significantly high nematicidal activity, this compound was found most toxic in the series, followed by compound 4-hydroxy-2*H*-1-benzopyran-2-one (**X**), which has also shown an appreciable activity of 80 % at 1000 ppm. Compound 7-hydroxy-4-methyl-2*H*-1-benzopyran-2-one (**VII**) was least active in the series. Rest of the compounds was moderately active. Irrespective of compounds, concentration 1000 ppm proved to be most toxic to nematode. Further nematicidal activity of these compounds was recorded after

48h. There was an increase in activity after 48 h, as in case of 4-hydroxy-2*H*-1-benzopyran-2-one (**X**); it increased from 80 to 90 % at 1000 ppm. Similarly, there was an overall increase in the nematicidal activity of rest of the compounds. In this series, all the compounds and concentration were statistically different from one another.

Various substituted 2-oxo-2*H*-1-benzopyran-7-yl/7,8-diyl/4-yl phenyl carbamates (**XII-XVI**) were also tested against *M. javanica* at 1000, 500, 250 and 125 ppm concentrations. All of these synthesized compounds were found highly toxic with 100 % mortality of nematodes at all tested concentrations. Introduction of carbamoyloxy moiety in 2*H*-1-benzopyran-2-one resulted into high nematicidal activity. Due to their high nematicidal activity, they were further evaluated at lower concentration *viz.* 120, 60, 30, 15 and 7.5 ppm and were found active even up to 7.5 ppm. Perusal of activity data revealed that 100 % mortality was found up to 7.5 ppm concentration. Mortality was recorded after 24 h only because no J_2 s revived after 48 h, clearly showing the highest activity of all compounds, irrespective of compounds and concentrations. All of these synthesized compounds were most toxic to nematodes with 100 % mortality at all tested concentrations. The compounds showed appreciable activity.

Conclusion

From the present studies, it may concluded that substituted 2*H*-1-benzopyran-2-ones serve as a potential lead compounds for the synthesis of various substituted 2-oxo-2*H*-1-benzopyran-7-yl/7,8-diyl/4-yl phenyl carbamates. The synthesized carbamates have exhibited a promising nematicidal activity. Compound 6-chloro-7-hydroxy-4-methyl-2*H*-1-benzopyran-2-one (**IX**) showed 100 % mortality at all tested concentrations and proved to be highly nematotoxic. Substituted 2-oxo-2*H*-1-benzopyran-7-yl/7,8-diyl/4-yl phenyl carbamate (**XII-XVI**) compounds were found highly toxic with 100 % mortality of nematodes at all tested concentrations. These compounds need further exploration for their possible use as nematocides and deserve further investigation.

TABLE-1
NEMATICIDAL ACTIVITY OF VARIOUS SUBSTITUTED 2*H*-1-BENZOPYRAN-2-ONES (VI-X)
AGAINST *Meloidogyne javanica* AFTER 24 h

Compound No.	Substituted 2 <i>H</i> -1-benzopyran-2-one			Mean of three replicates
	% J_2 mortality after 24 h				
	R ₁	R ₂	R ₃	1000 ppm	500 ppm	250 ppm	125 ppm	Mean (compd.)
VI	H	H	CH ₃	31.0 (33.8)	20.0 (26.5)	9.7 (18.1)	0.0 (0.0)	15.2 (19.6)
VII	H	H	H	26.5 (30.9)	12.7 (20.8)	0.0 (0.0)	0.0 (0.0)	9.8 (12.9)
VIII	OH	H	H	50.0 (45.0)	40.0 (39.1)	23.3 (28.8)	20.0 (26.1)	33.3 (34.7)
IX	H	Cl	H	100.0 (90.0)	100.0 (90.0)	100.0 (90.0)	100.0 (90.0)	100.0 (90.0)
X	4-hydroxy 2 <i>H</i> -1-benzopyran-2-one			80.3 (63.6)	64.5 (53.4)	38.7 (38.4)	24.1 (29.3)	51.9 (46.2)
	Mean (Conc.)			57.5 (52.7)	47.4 (46.0)	34.4 (35.1)	28.8 (29.1)	—

C.D (5 %): Compound = (2.12); Concentration = (1.89); Compound x Concentration = (4.24)
Values in parentheses are angular transformed value.

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REFERENCES

1. P. Selvam, N. Ramlakshmi and S.K. Kumar, *Int. J. Pharm. Sci. Res.*, **2**, 413 (2011).
2. K. Takaishi, M. Izumi, N. Baba, L. Kawazu and S. Nakajima, *Bioorg. Med. Chem. Lett.*, **18**, 5614 (2008).
3. R.G. Kalkhambkar, G.M. Kulkarni, C.M. Kamanavalli, N. Premkumar, S.M.B. Asdaq and C.M. Sun, *Eur. J. Med. Chem.*, **43**, 2178 (2008).
4. R.L. Baron, *Handbook of Pesticide Toxicology*, Academic Press, New York, pp. 1125-1189 (1991).
5. S. Perveen, N. Fatima, K.M. Khan, A. Khan, M. Ali and M.I. Choudhary, *J. Chem. Soc. Pak.*, **32**, 338 (2010).
6. J. Yoshida, S. Kuroiwa, M. Sasaki and F. Hirata, *Nippon Kagakkai Koenyokoshu*, **82**, 188 (2002).
7. T.W. Greene and P.G.M. Wuts, *Protective Groups in Organic Synthesis*, Wiley & Sons, New York, pp. 309-348 (1991).
8. J.R. Corbett, K. Wright and A.C. Baillie, *The Biochemical Mode of Action of Pesticides*, Academic Press, London, edn 2, pp. 382 (1984).
9. R. Singh, T. Batra, M.S. Malik and O.P. Malik, *Chim. Acta Turc.*, **23**, 103 (1995).
10. I. Chopra, R.K. Walia and R. Singh, *Pestic. Res. J.*, **18**, 124 (2006).
11. R. Singh, B.B. Gupta, O.P. Malik and H.R. Kataria, *Pestic. Sci.*, **20**, 125 (1987).
12. K.M. Khan, Z.S. Saify, S. Begum, F. Noor, M.Z. Khan, S. Hayat, M.I. Choudhary, S. Perveen, Atta-Ur-Rahman and Zia-Ullah, *Nat. Prod. Res.*, **17**, 115 (2003).
13. B.S. Furniss, A.J. Hannaford, V. Rogers, P.W. Smith and A.R. Tatchell, *Text Book of Practical Organic Chemistry (Vogel)*, Great Britain ELBS and Longmann, edn 4, p. 925 (1978).
14. E.V. Stoyanov and J. Mezger, *Molecules*, **10**, 762 (2005).
15. S.J. Park, J.C. Lee and K.I. Lee, *Bull. Korean Chem. Soc.*, **28**, 1203 (2007).
16. K. Gill, S.K. Mehta, M.S. Malik, O.P. Malik and R.K. Walia, *Nematol. Mediterr.*, **29**, 219 (2001).