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D-phenyl alanine-oxalic acid (*D*-Phe-Ox): A novel nano ionic liquid catalyzing the one-step green synthesis of functionalized spirolactones and dispirodihydrofuranyl oxindoles

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ABSTRACT

In this paper, a new nano-size ionic liquid (nIL) has been synthesized from *D*-phenylalanine and oxalic acid (*D*-Phe-OX) *via* a simple procedure. The obtained nIL has been characterized by the fourier transform infrared spectroscopy (FT-IR), energy-dispersive X-ray spectroscopy (EDAX), thermogravimetric analysis (TGA), nuclear magnetic resonance spectroscopy (^{1}H NMR), and field emission scanning electron microscopy (FESEM), techniques. The synthesized nanocatalyst has been used for the three-component one-step synthesis of functionalized spirolactones *via* the condensation reaction of arylamines, DMAD and isatins. The stereoselective preparation of dispirodihydrofuranyl oxindoles has also been examined successfully through the one-step pseudo-four component condensation of isatins, amines and DMAD, respectively.

Keywords: *Nano ionic liquid, DMAD, Spirolactone, Dispirodihydrofuranyl oxindole, Chiral, Isatin.*

1. Introduction

Ionic liquids are compounds which consist of bulky organic cations and inorganic/ organic anions, which melt at a relatively low temperature. They are inflammable, thermally stable, easy and safe solids to handle with negligible vapor pressure and soluble in many polar organic molecules and also water. Their polarity and hydrophilicity/ hydrophobicity depend on the suitable combination of cations and anions. They are known by several different names such as neoteric solvents, ionic fluids, and melting salts and usually could be utilized as both solvent and catalyst which is in synchronized with green chemistry rules [1].

Spiro compounds represent an important class of naturally occurring substances identified by highly pronounced biological properties. Some spirocyclic oxindole systems demonstrated antimicrobial [2], antibacterial [3], and antitumor [4] properties. Some alkaloids including spirooxindole motifs have been discovered [5].

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Recently, versatile efficient methods have been reported for the preparation of spirooxindole-fused heterocycles such as cycloaddition [6], Asinger-type reaction [7], domino Buchwald–Hartwig/ Michael reaction [8], cascade cyclization and Friedel-Crafts reaction [9], and Morita-Baylis-Hillman [10] reactions.

Isatin derivatives are of the critical precursors for preparation of a wide range of spirocyclic oxindoles [11,12]. Spiro compounds consist of lactone motif has appeared recent years because of their different pharmaceutical and biological properties such as anti-androgenic [13], anti-convulants [14], anti-HIV [15], and anti-inflammatory [16] activities.

In continuation of our research interest in preparing nitrogen-containing heterocycles [17-22] herein we report synthesis of functionalized spirolactones and dispirodihydrofuranyl oxindoles *via* the reactions of isatin, primary amines, and DMAD. The transformation has been accelerated in the presence of *D*-phenylalanine and oxalic acid (*D*-Phe-OX), as a novel, newly reported chiral nano-size ionic liquid. The IL has also been characterized by ordinary techniques. It must be mentioned that some derivatives of these compounds have been obtained by Perumal et al. in 2012 in MeOH

[23]. In our report the stereoselectivity of the dispirodihydrofuranyl oxindoles and the reaction mechanisms have been investigated.

2. Experimental

2.1. Reagents and apparatus

All chemicals were purchased from Merck, Aldrich, and Alfa Aesar and were used without further purification. IR spectra were recorded from KBr disk using the FT-IR Bruker Tensor 27 instrument. Melting points were determined on an Electrothermal 9200 analyzer and are uncorrected. ^{1}H NMR spectra were recorded with a Bruker drx 400 MHz, machine. Elemental analyses were determined using a Thermo-Finnigan Flash EA 1112 Series. Mass spectroscopy has been obtained by Gc-Mass 5973 network mass selective detector, GC 6690 Agilent device. Progress of the reaction was monitored by thin layer chromatography TLC technique using commercially available silica gel sheets. Preparative layer chromatography (PLC), carried out on 20 × 20 cm^2 plates, coated with a 1 mm thick layer of Merck silica gel PF_{254}, prepared by applying the silica as slurry and drying in air. The scanning electron microscope FESEM, model ZEISS AMA, was used to characterize the nanostructures. Thermogravimetric analysis was determined by PYRIS DIAMOND apparatus. The polarimeteric machine SCHMIDT, HAENDCH 23368, was used to characterize optical rotation. The products were characterized by comparison of their melting points and also spectroscopic data (FT-IR, ^{1}H NMR, and CHN analysis).

2.2. General procedure for the synthesis of D-phenylalanine-Oxalic acid nano ionic liquid (D-Phe-OX nIL)

A solution of *D*-phenylalanine (4 mmol, 0.66 g), and oxalic acid dihydrate (4 mmol, 0.504 g), in 5 ml water was stirred at 90 °C for 2 h. Then, solvent was evaporated to give a snow-like white powder.

2.3. General procedure for the synthesis of spirolactones (4a-h) and dispirodihydrofuranyl bisoxindoles (4a-h)

A mixture of isatins (**1a-c**, 1 mmol for spirolactones and 2 mmol for dispirodihydrofuranyl bisoxindoles), arylamines **(2a-g**, 1 mmol), DMAD (**3**, 1 mmol), and nIL 0.008 g, under solvent-free conditions at 70 °C was stirred for the appropriate reaction time monitored by thin layer chromatography TLC (*n*-hexan/ EtOAc eluent, 3:1). After completion of the reaction, the pure products **4a-h** and **5a-h** were gained by plate chromatography.

Selected spectral data

*Methyl-5'-nitro-2',5-dioxo-4-(phenylamino)-5H-spiro[furan-2,3'-indoline]-3-carboxylate **(4g)**:*

Orange solid. m.p.: 107-110 °C. IR (KBr): $\bar{\nu}$ = 3617, 3285, 3100, 2953, 1739, 1621, 1462, 1528, 1340, 1208, 1153 cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 3.72 (s, 3H, -OCH_3), 6.99-8.4 (m, -ArH), 10.92 (s, 1H, NH), 11.69 (s, 1H, NH) ppm. GC-Mass: m/z= 378 ([M$^+$-2]-Me), 314 (M$^+$-CH_2CHCO_2Me), 302 ([M$^+$]-aniline), 284 ([M$^+$]-CO_2, -NHCOEt), 270 ([M$^+$]-$C_6H_5NO_2$). Anal. calcd. for $C_{19}H_{13}N_3O_7$: C, 57.73; H, 3.31; N, 10.63%. Found: C, 57.74; H, 3.30; N, 10.64%.

*Methyl-5'-chloro-2',5-dioxo-4-(p-tolylamino)-5H-spiro[furan-2,3'-indoline]-3-carboxylate (**4h**):*

Dark yellow solid. m.p.= 85-90 °C. IR (KBr): $\bar{\nu}$ = 3445, 2924, 1741, 1660, 1466, 1249, 1169, 671 cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 2.23 (s, 3H, -CH_3), 3.56 (s, 3H, -OCH_3), 6.61-7.96 (m, -ArH), 9.99 (s, 1H, NH), 10.33 (s, 1H, NH) ppm. GC-Mass: m/z= 372 ([M$^+$]-CO), 344 ([M$^+$]-CO, -CH_2CH_2), 328 ([M$^+$]-CO, -CH_2CH_2, -Me), 316 ([M$^+$]-CH_2CHCO_2Me), 298 ([M+]-CH_2CHCO_2Me, -Me), 227 ([M$^+$]-4-ClC_6H_4NHCHO), 194 ([M$^+$]-CO_2, -NH_2COMe, -4-ClC_6H_5). Anal. calcd. for $C_{20}H_{15}N_2O_5Cl$: C, 60.23; H, 3.79; N, 7.02%. Found: C, 60.24; H, 3.80; N, 7.01%.

*Dispirodihydrofuranyl bisoxindole (**5g**):*

Orange solid. m.p.= 135-138 °C. IR (KBr): $\bar{\nu}$ = 3328, 3130, 2954, 1726, 1620, 1460, 1528, 1341, 1251, 1160 cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 3.15 (s, 3H, -OCH_3), 6.93-8.57 (m, -ArH), 9.16 (s, 1H, NH), 10.47 (s, 1H, NH), 10.94 (s, 1H, NH) ppm. Anal. calcd. for $C_{26}H_{17}N_5O_9$: C, 57.46; H, 3.15; N, 12.89%. Found: C, 57.45; H, 3.16; N, 12.90%.

*Dispirodihydrofuranyl bisoxindole (**5h**):*

Dark yellow solid. m.p.= 90-94 °C. IR (KBr): $\bar{\nu}$ = 3360, 2953, 1736, 1616, 1436, 1575, 1249, 1171, 651 cm^{-1}. ^{1}HNMR (DMSO-d_6, 400 MHz): δ= 2.23 (s, 3H, -CH_3), 3.76 (s, 3H, -OCH_3), 6.55-7.58 (m, -ArH), 10.16 (s, 1H, NH), 10.19 (s, 1H, NH), 11.16 (s, 1H, NH) ppm. GC-Mass: m/z= 372 ([M$^+$]-4-chlorooxindolyl), 357 ([M$^+$]-4-chlorooxindolyl, -Me), 328 ([M$^+$]-4-chlorooxindolyl, -Me, -CO), 265 ([M$^+$]-4-chlorooxindolyl, -4-methylaniline). Anal. calcd. for $C_{27}H_{19}N_3O_5Cl_2$: C, 65.19; H, 3.85; N, 8.45%. Found: C, 65.20; H, 3.86; N, 8.43%.

3. Results and Discussion

The chiral ionic liquid (*D*-Phe-OX), a snow-like white powder, was prepared *via* a simple procedure through the reaction of *D*-phenylalanine and oxalic acid dihydrate in the presence of water as a solvent (Scheme 1).

The structures of catalyst were consistent with FT-IR, FESEM, EDAX, ^{1}HNMR, TGA/ DTG, and polarimetric techniques. The FT-IR spectrum of *D*-Phe-OX, exhibited sharp stretching peaks at 1698 with a shoulder at 1741 cm^{-1} related to carboxylic acid and carboxylate carbonyl groups. The sharp peaks at 1264, 1131, 514, 3061, and 2930 cm^{-1} dedicated the C-O stretching of oxalate ion, C-OH stretching vibration of carboxylic acid, rotational vibration of NH^{3+}, sp^2 C-H, and sp^3 C-H stretching bands respectively. The broad starching peaks in 3862-3898 cm^{-1} and 2600-3100 cm^{-1} region demonstrated N-H stretching and NH^{+3} vibrations (Fig. 1).

^{1}H NMR spectrum of *D*-Phe-OX, which is recorded in D_2O, showed two doublets of doublet at δ 3.07 J = 13.39, 10.91 Hz, and 3.23 ppm J = 19.39, 7.35 Hz, relating to the CH_2 methylene group. The CH proton of *D*-phenylalanine appeared at 4.17 ppm as a doublet of doublet pattern (J = 10.19, 7.46 Hz). Aromatic protons were recorded at two multiple peaks at δ 7.19-7.22 ppm 2H, and 7.25-7.34 ppm 3H (Fig. 2). The protons of carboxylic acid and NH^{3+} groups did not appear due to exchanging with the deuterated solvent.

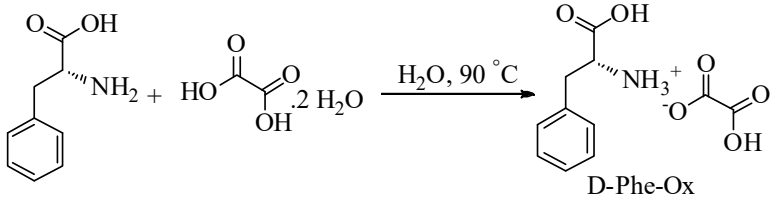
The TGA/ DTG diagram of the nano IL is shown at Fig. 3. According to the results, the weight loss took place in some exothermic steps which consist of 50-70 °C and 120-140 °C. These peaks are due to losing the adsorbed water on the surface of IL. The main weight loss was displayed at 210-230 °C, the complete destruction of structure was related to decomposition of *D*-phenylalanine happening at 300-380 °C.

The FESEM pattern of synthesized catalyst is shown at Fig. 4. This image reveals that the product consists of nano-size parts with the average size of 40-75 nm, with semi-filamentary morphology.

EDAX of *D*-Phe-OX nIL in Fig. 5 demonstrated that the catalyst is only made up of carbon, nitrogen, and oxygen.

The optical rotation for the synthesized nIL by means of polarimeter device obtained ($[\alpha_o]_D^{25} = 18.25$ 25 °C, C = 4×10^{-4} g/ml, λ_{Na}), whereas in the same conditions the optical rotation for D-phenylalanine is $[\alpha_o]_D^{25} = 55.25$. these results confirmed the chirality of *D*-Phe-OX.

In order to examine the catalytic activity of the prepared nIL, our present investigation deals with the reactions of isatin, primary amines, and DMAD in the presence of chiral nano ionic liquid, *D*-Phe-OX, under solvent-free conditions to obtain functionalized spirolactones (Scheme 2).



Scheme 1. Preparation of *D*-Phe-OX nano IL.

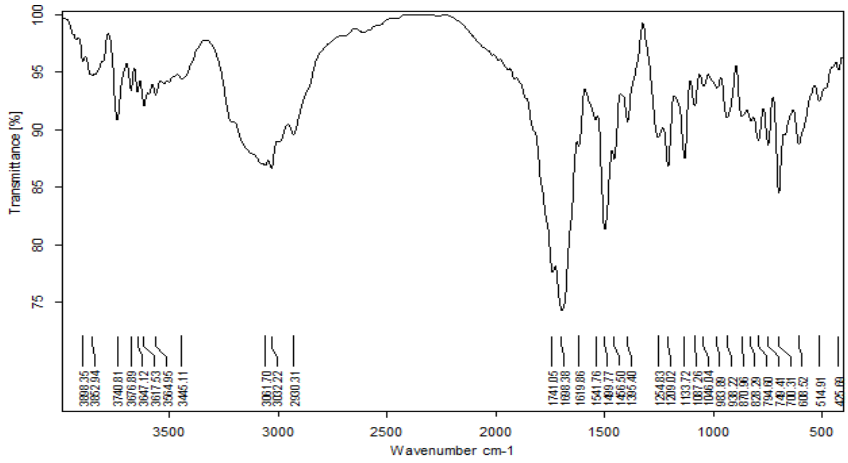


Fig. 1. FT-IR spectrum of *D*-Phe-OX.

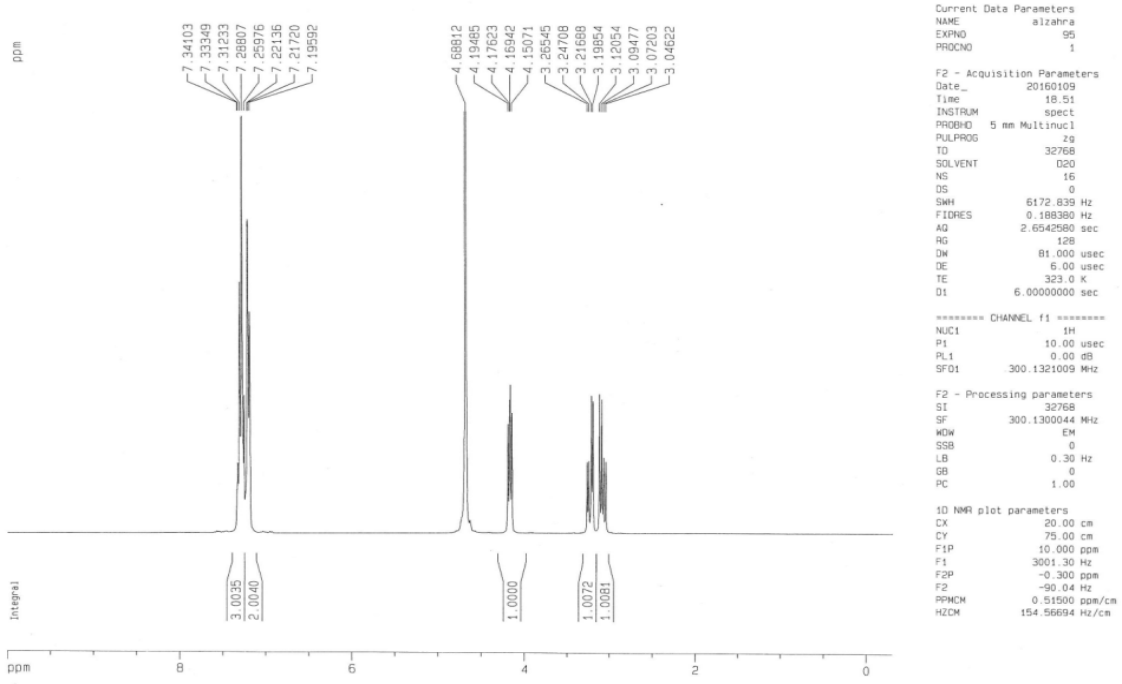


Fig. 2. [1]HNMR spectrum for *D*-Phe-OX in D_2O.

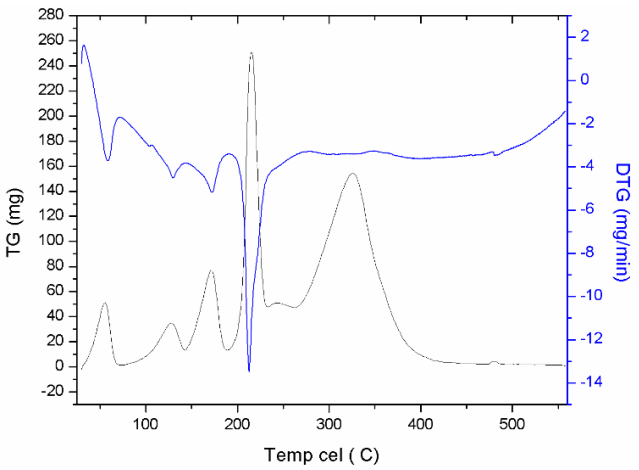


Fig. 3. TGA/ DTG diagram of *D*-Phe-OX.

To optimize the reaction conditions, the one-pot and one-step reaction of isatin (**1a**, 1 mmol), 4-bromoanilin (**2b**, 1 mmol), and DMAD (**3**, 1 mmol), in the presence of D-Phe-OX was picked as the model reaction. The effect of various factors such as temperature, solvent, and catalyst amount has been checked. The results Table 1, showed that the best temperature is 70 °C (entries 1-3). Increasing up to 90 °C didn't have any significant effect on the reaction progress (entry 4). The solvent investigation in entries 5-8, confirmed that the reaction performed well in the absence of solvent.

The catalyst amount, as another item, has also been examined which resulted in 0.008 g of nIL (entries 9,10). The model reaction has also surveyed *via* a domino manner but the corresponding product **4b** was not obtain.

In the next step, based on optimized conditions, the reaction of various isatins (**1a-c**) with aniline derivatives (**2a-g**), and DMAD (**3**) in 1:1:1 molar ratio proceeded in the presence of catalytic amount *D*-Phe-OX nIL (0.008 g) under solvent-free conditions at 70 °C. The results are summarized in Table 2. As could be deduced, the reaction of aniline (**2a**) with isatin (**1a**) progressed in a longer time in comparison to the condensation of **2a** with electron withdrawing 4-nitroaniline (**1c**). In addition, condensation of electron-donating anilines performed in longer period in respect of electron-withdrawing analogues.

We have expressed a plausible mechanism for the formation of spirolactone. First, Bronsted acidic sites of nIL activated DMAD (**3**), which was condensed with arylamine (**2**) to give intermediate (**A**) in the presence of *D*-Phe-OX nIL as the Bronsted acid catalyst. Second, the intermediate (**A**) attacked its carbanionic moiety to activated isatin (**1**) to produce **B**. Finally, the spirolactone compounds (**4**) were obtained through intramolecular cyclization followed by methanol removal form **B** (Scheme 3).

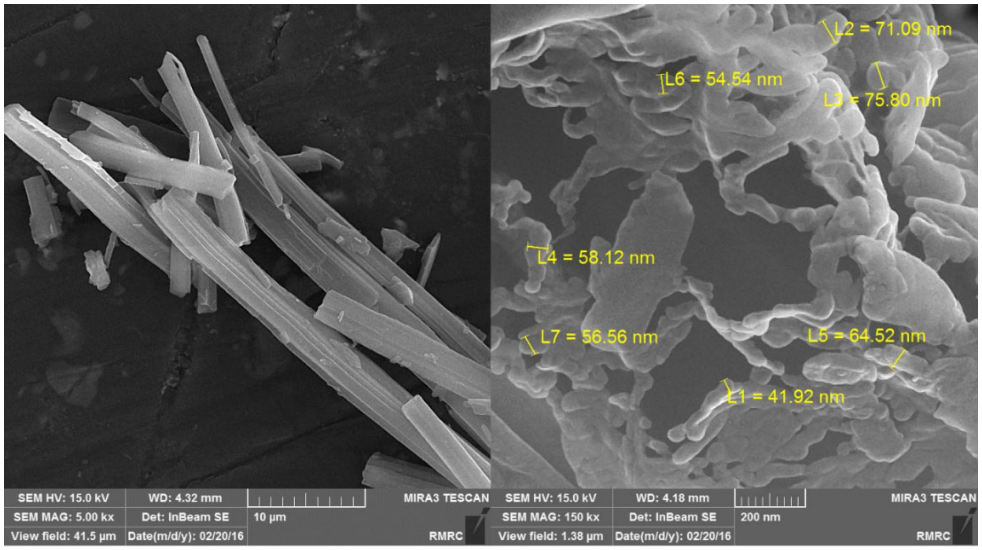


Fig. 4. FESEM image of nano-size *D*-Phe-OX.

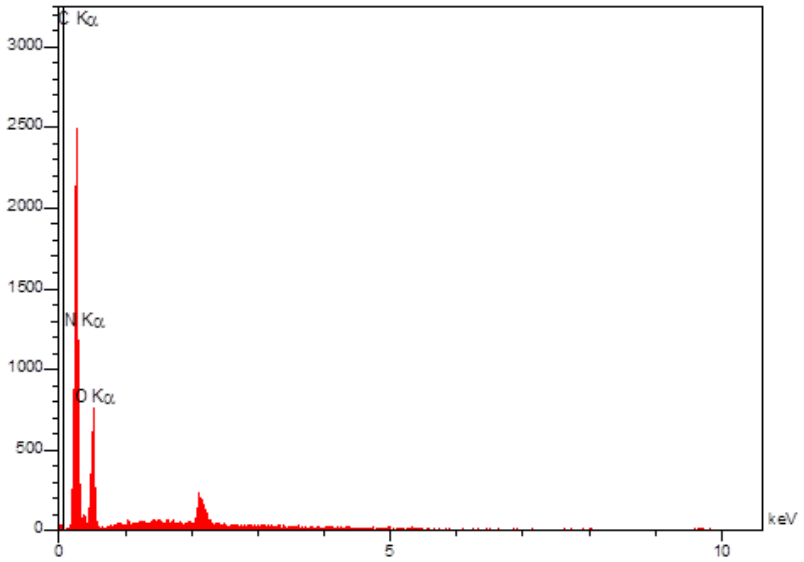
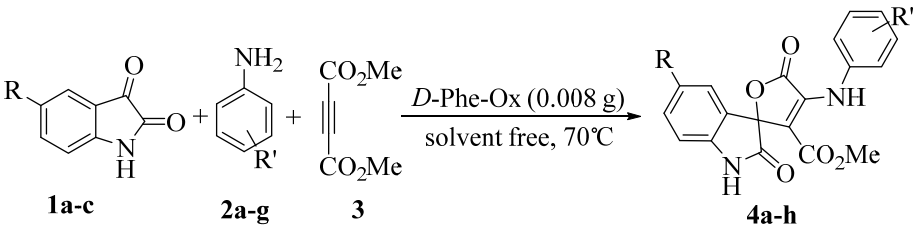


Fig. 5. EDAX diagram of *D*-Phe-OX nIL.



1a-c

a R = H
b R = NO_2
c R = Cl

2a-g

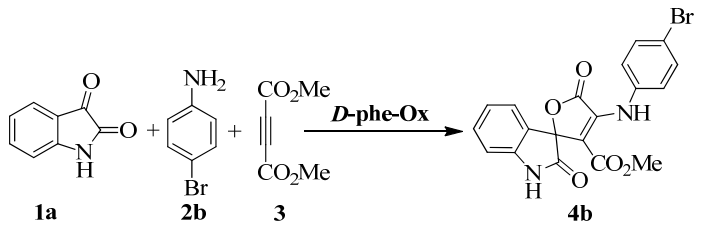
a R' = H
b R' = 4-Br
c R' = 3-Me
d R' = 4-Cl
e R' = 4-NO_2
f R' = 4-OMe
g R' = 4-Me

3

4a-h

Scheme 2. Synthesis of spirolactones using *D*-Phe-OX.

Table 1. Rationalization of the reaction conditions for the synthesis of **4b.**



Entry	Solvent[a]/ Temp (°C)/ nIL amount (g)	Time (min)	Yield (%)[b]
1	-/ rt/ 0.008	360	90
2	-/ 50/ 0.008	60	70
3	-/ 70/ 0.008	35	90
4	-/ 90/ 0.008	35	90
5	CH_3OH/ 70/ 0.008	150	50
6	CH_3CN/ 70/ 0.008	75	60
7	H_2O/ 70/ 0.008	135	10
8	C_2H_5OH/ 70/ 0.008	135	20
9	-/ 70/ 0.004	55	90
10	-/ 70/ 0.012	40	90

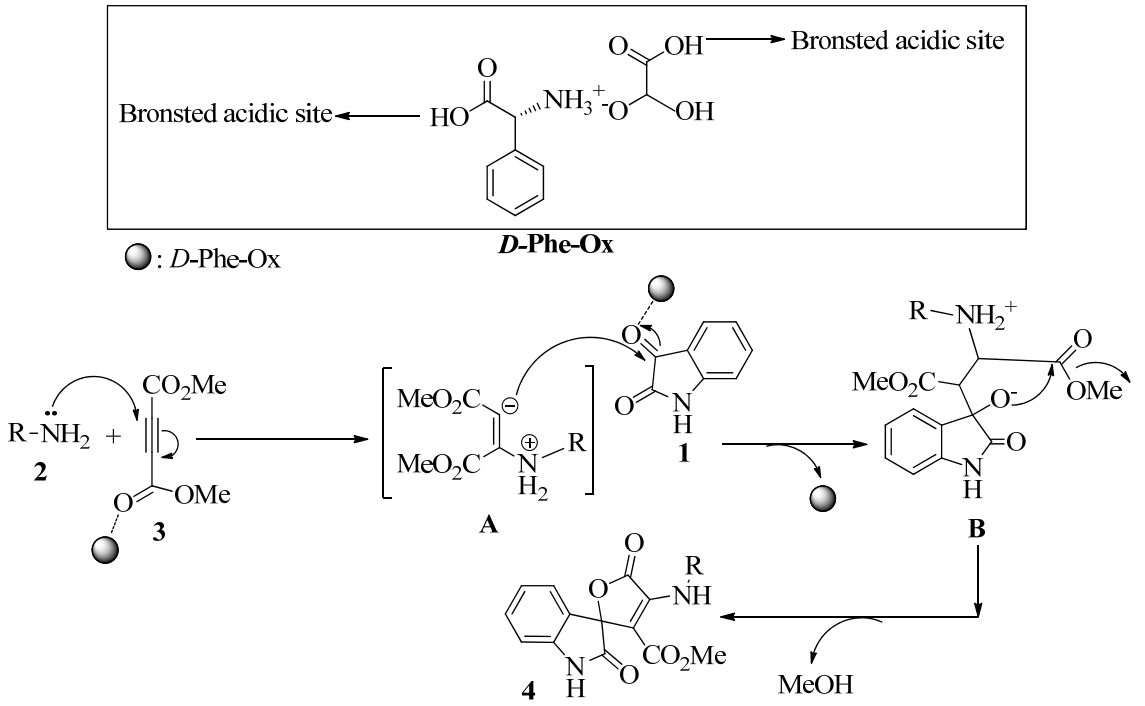
[a]5 ml of each solvent has been used.
[b]Isolated yield.

Table 2. Synthesis of spirolactone derivatives in the presence of D-Phe-OX nIL.

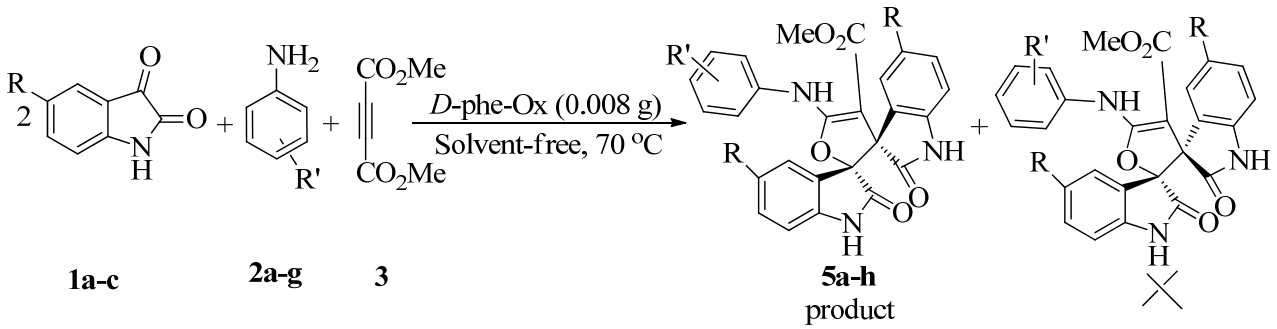
Entry	R	R'	Product	Time (min)	Yield (%)[a]
1	H	H	**4a**	55	92
2	H	4-Br	**4b**	35	90
3	H	3-CH_3	**4c**	100	91
4	H	4-Cl	**4d**	30	90
5	H	4-NO_2	**4e**	60	73
6	H	4-OCH_3	**4f**	70	81
7[b]	NO_2	H	**4g**	30	94
8[b]	Cl	4-CH_3	**4h**	150	95

[a]Isolated yield.
[b]New compounds.

In the next part of our research, the synthesis of dispirodihydrofuranyl oxindoles has been examined via a one-pot, one-step and pseudo-four-component condensation reaction of arylamines, DMAD, and isatins, in 1:1:2 molar ratio, in the presence of 0.008 g of the nIL under solvent-free conditions at 70 °C (Scheme 4). Based on the results, only one diastereomer of the corresponding product has been obtained. This diastereoselectivity was because of utilizing *D*-Phe-OX, as a chiral catalyst. Observation of one spot in TLC and ^{1}HNMR analysis and also plate chromatography isolation prove this claim. The assumed route for the synthesis of dispirodihydrofuranyl bisoxindoles has been demonstrated in Scheme 5. The intermediate (**C**) has been prepared according to Scheme 3, which releases CO_2 *via* decyclization process to produce (**D**). Then, the zwitterionic compound (**D**) was attached to the second activated isatin molecule to create dispirodihydrofuranyl bisoxindoles (**5**) diastereoselectively.



Scheme 3. Possible mechanism for the synthesis of spirolactones using *D*-Phe-OX nIL.



Scheme 4. One-pot synthesis of dispirodihydrofuranyl bisoxindoles in the presence of *D*-Phe-OX nIL.

It is also noteworthy to mention that this is the first report on the synthesis of diastereoselective dispirodihydrofuranyl oxindoles using a chiral nano ionic liquid. In regard to the obtained results in Table 3, formation of products in presence of 5-nitroisatin (**1b**) in comparison to 5-chloroisatin (**1c**) was performed in shorter time. Likewise, the condensation of electron-donating analogues of aniline, such as 4-methoxyaniline (**2f**) and 4-methylaniline (**2g**) promoted more rapidly in shorter time as opposed to aniline (**2a**) and its electron-withdrawing analogues such as 4-chloroaniline (**2d**), and 4-nitroaniline (**2e**).

4. Conclusions

In conclusion in this report, a novel nano-size chiral ionic liquid has been prepared for the first time and characterized *via* FESEM, FT-IR, EDX, TGA/ DTG, and ^{1}H NMR techniques. Its catalytic activity has been examined in the synthesis of spirolactones and dispirodihydrofuranyl bisoxindoles under solvent-free conditions successfully. This work has several advantages such as short reaction times, high yields of products, ambient temperature; and utilizing easily prepared, inexpensive and non-toxic nano catalysts. The procedure has not required hazardous organic solvents, which follows the green chemistry. The steresoselectivity of dispirodihydrofuranyl bisoxindoles synthesis has also been studied.

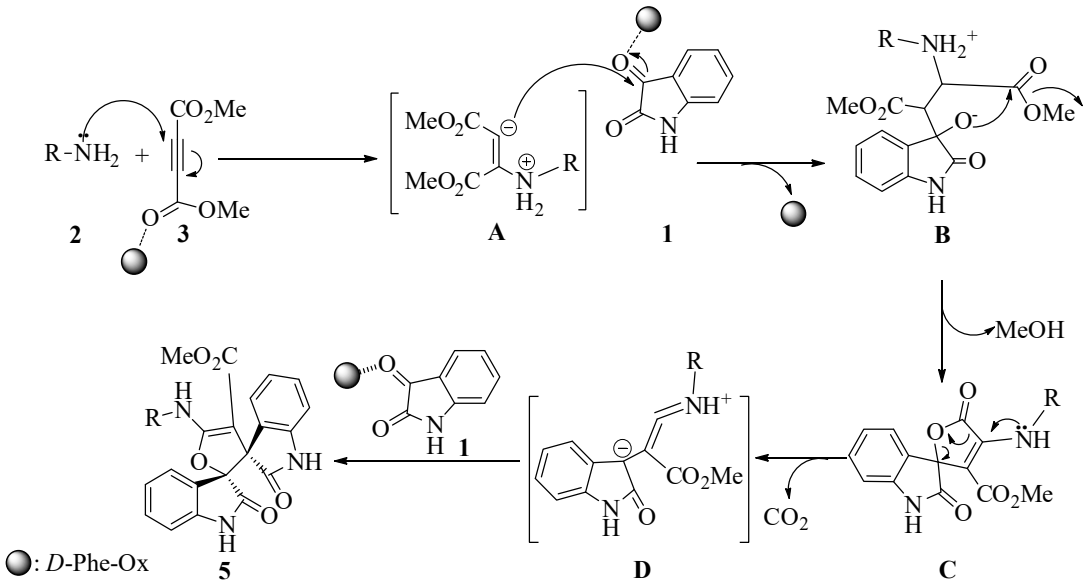
Acknowledgments

The authors express their gratitude for financial support from Alzahra University.

Table 3. Synthesis of dispirodihydrofuranyl bisoxindole derivatives in the presence of D-Phe-OX nIL.

Entry	R	R'	Product	Time (min)	Yield (%)[a]
1	H	H	**5a**	240	92
2	H	4-Br	**5b**	180	91
3	H	3-CH_3	**5c**	60	85
4	H	4-Cl	**5d**	210	93
5	H	4-NO_2	**5e**	330	85
6	H	4-OCH_3	**5f**	90	92
7[b]	NO_2	H	**5g**	85	93
8[b]	Cl	4-CH_3	**5h**	120	94

[a]Isolated yield.
[b]New compounds.



Scheme 5. Proposal mechanism for the synthesis of dispirodihydrofuranyl bisoxindoles using *D*-Phe-OX nIL.

References

[1] a) R. Skoda-Flodes, Molecules 19 (2014) 8840-8884. b) C. Chiappe, D. Pieraccini, J. Phys. Org. Chem. 18 (2005) 275-297. c) T.L. Greaves, C. Drummond, Chem. Rev. 115 (2015) 11379-11448.

[2] A. Dandia, Sh. Khan, P. Soni, A. Indora, D.K. Mahawar, P. Pandy, C.S. Chauhan, Bioorg. Med. Chem. Lett. 27 (2017) 2873-2880.

[3] P. Ramesh, Rao K.S., R. Trivedi, B.S. Kumar, R.S. Prakasham, B. Sridhar, RSC Adv. 6 (2016) 26546-26552.

[4] S.A.S. Ghozlan, M.F. Mohamed, A.G. Ahmed, S.A. Shouman, Y.M. Attia, I.A. Abdelhamid, Arch. Pharm. 348 (2015) 113-124.

[5] G.O. Fonseca, J.M. Cook, Org. Chem. Ins. 6 (2016) 1-55.

[6] J. Sun, H. Gong, Ch. Yan, Chin. J. Chem. 33 (2015) 1049-1056.

[7] G. Rainoldi, F. Begnini, A. Silvani, G. Lesma, Synlett 27 (2016) 2831-2835.

[8] N. Sharma, Zh. Li, U.K. Sharma, E.V. Van der Eycken, Org. Lett. 16 (2014) 3884-3887.

[9] U. Borthakur, M. Borah, M.J. Deka, A.K. Saikia, J. Org. Chem. 81 (2016) 8736-8743.

[10] H.P. Deng, Y. Wei, M. Shi, Org. Lett. 13 (2011) 3348-3351.

[11] a) Gh. Mohammadi Ziarani, R. Moradi, N. Lashgari, Tetrahedron 74 (2018) 1323-1353. b) W.B. Chen, Z.J. Wu, Q.L. Pei, N.F. Cun, X.M. Zhang, W.C. Yuan, Org. Lett. 12 (2010) 3132-3135.

[12] a) L. Hong, R. Wang, Adv. Synth. Catal. 355 (2013) 1023-1052. b) M. Bai, B. Cui, J. Zuo, J. Zhao, Y. You, Y. Chen, X. Xu, X. Zhang, W. Yuan, Tetrahedron 71 (2015) 949-955.

[13] M. Messina, C. Manieri, P. Biffignandi, C. Massucchetti, R.F. Novi, G.M. Molinatti, J. Endocrinol. Invest. 6 (1983) 23-27.

[14] E.M. Peterson, K. Xu, K.D. Holland, A.C. McKeon, S.M. Rothman, J.A. Ferrendelli, D.F. Covey, J. Med. Chem. 37 (1994) 275-280.
[15] T. Hayashi, M. Jean, H. Huang, S. Simpson, N.G. Santoso, J. Zhu, Antivir. Res. 146 (2017) 76-85.
[16] L. Yang, L.H. Qin, S.W. Bligh, A. Bashall, C.F. Zhang, M. Zhang, Z.T. Wang, L.S. Xu, Bioorg. Med. Chem. 15 (2006) 3496-3501.
[17] K. Nikoofar, Z. Khademi, M. Haghighi, J. Chem. Sci. 128 (2016) 1805-1811.
[18] L. Fotouhi, K. Nikoofar, Tetrahedron Lett. 54 (2013) 2903-2905.
[19] K. Nikoofar, Sh. Moazzez Dizgarani, Monatsh. Chem. 146 (2015) 1161-1204.
[20] K. Nikoofar, Z. Khalili, Z.Z. Naturforsch. B Chem. Sci. 71 (2016) 31-36.
[21] K. Nikoofar, S. Gorji, Phosphorus Sulfur Silicon Relat. Elem. 190 (2015) 1138-1145.
[22] K. Nikoofar, M. Haghighi, Z. Khademi, Arab. J. Chem. (2016) doi:10.1016/j.arabjc.2016.01.013.
[23] S.E. Kiruthika, R. Amritha, P.T. Perumal, Tetrahedron Lett. 53 (2012) 3268-3273.