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INTRODUCING A NEW METHODOLOGY OF ALDEHYDE SYNTHESIS FROM ALKYL HALIDE USING α-CHLORO NITRONE AS A NEW, STABLE AND POTENTIAL OXIDIZING REAGENT

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ABSTRACT

Consecutive S_N2 reaction of α-chloro nitrones are studied with alkyl halides and the nitrones are found to have remarkable oxidizing properties for the conversion of alkyl halides to aldehydes with high yield. In addition, the side product obtained can serve as efficient dipolarophile in 1,3 DCR to produce spiro cycloadducts in good yields.
Keywords: α-chloro nitrone as oxidizing reagent, S_N2 reaction, aldehyde synthesis, spiro cycloadduct

INTRODUCTION

Convertion of alkyl halides to aldehydes using *N*-oxide with moderate yields have been already reported (Krohnke reaction). In addition to the existing methods available for the synthesis of aldehyde from alkyl halides,[1-6] we would like to incorporate an efficient one pot synthesis of aldehyde from alkyl halides using for the first time α-chloro nitrones (**1**) as a new, stable and potential oxidizing reagent with an excellent yield (Scheme-1, Table 1). In addition, the side product (furan derivatives, **2**) obtained during aldehyde synthesis has been successfully used as dipolarophile in 1,3-DCR with nitrone (**1**) for the production of spiro cycloadducts (**3**) with high yields (almost **75 – 85%** ; Scheme-2). α-chloro nitrones (**1**) are more reactive than other nitrones due to the electron withdrawing effect of chlorine and therefore can act as more powerful oxidizing agent than other nitrones.

Literature survey reveals that aldehyde synthesis using nitrone as active oxidizing reagent and further use of side products (obtained during aldehyde synthesis) as dipolarophile in cycloaddition reactions are not yet known and hence can be incorporated as an important application in nitrone chemistry. Synthesis and 1,3 dipolar cycloaddition reactions of *N*-phenyl-α-chloro nitrone[7,8](**1**, R=Ph) has been already reported. Following the same methodology, novel *N*-methyl-α-chloro nitrone (**1**,R=Me) has been synthesized as white crystalline solid, m.p 52^0C (uncorrected) and used for aldehyde synthesis as oxidizing reagent.

EXPERIMENTAL

General remarks :

^{1}H NMR spectra were recorded with a Bruker Avance DPX 400 spectrometer (400 MHz, FT NMR) using TMS as internal standard. ^{13}C NMR spectra were recorded on the same instrument at 100 MHz. The coupling constants (*J*) are given in Hz. IR spectra were obtained with a Perkin-Elmer RX 1-881 machine as film or KBr pellets for all the products. MS spectra were recorded with a Jeol SX-102 (FAB) instrument. The HRMS spectra were recorded on a Q–Tof micro instrument (YA–105). TLC was carried out on Fluka silica gel TLC cards while column chromatography was performed with silica gel (E.Merck India) 60–200 mesh. All other reagents and solvents were purified after receiving from commercial suppliers. *N*-methylhydroxylamine was purchased from Aldrich Chemical Company and was used as received. *N*-phenylhydroxylamine was prepared following standard methods available in literature and has been used in synthesis[9,10,11].

General procedure for the synthesis of nitrone 1 (R = Me)

N-methylhydroxylamine (250mg, 5.3127 mmole) was added to chlorohydrin (720mg,1 equivalent) in dry ether (50 mL) and anhydrous $MgSO_4$. The reaction mixture was kept at RT with constant stirring with a magnetic stirrer under N_2 atmosphere for 10 hr. The formation of nitrone was monitored by TLC (R_f = 0.34). The nitrone was isolated under reduced pressure vaccum pump as white niddle shape crystals (920mg, 94%; m.p: 52^0C)**.**

Spectroscopic data for nitrone 1 (R = Me)

Yield: 920mg (94%); white niddle shape crystals; R_f = 0.43, m.p: 52^0C (uncorrected); IR (KBr): 3595 – 3470 (br), 1660(s), 1610(s), 1415 (m), 1185 (s) cm^{-1}; ^{1}H NMR ($CDCl_3$): δ 5.84 (d, 1H, CH=N^+), 5.79 (br,1H, -OH, exchanged in D_2O), 3.51 (dd, 1H, *J* = 6.16, 6.08 Hz, CHCl), 3.31 (s, 3H, N^+- CH_3), 1.88 - 1.15 (m, 6H, CH_2 protons); ^{13}C NMR($CDCl_3$) : δ 141.55 (CH=N^+), 55.76 (CHCl), 34.84 (N^+- CH_3), 28.50, 27.22, 26.00 (3 CH_2 carbons); HRMS – EI: Calcd. for $C_6H_{12}O_2NCl$, (M), 165.5710, Found: M^+, 165.5698.

General procedure for synthesis of aldehyde (benzaldehyde) and furan derivative 2 (entry 1;Table 1)

To a stirred solution of nitrone **1** (R=Me; 500mg, 3.0198 mmol) in dry ether (25 ml) was added pyridine (1 equivalent) and stirred at RT with a magnetic stirrer under N_2 atmosphere for 1 hr while the formation of transient nitrone **1a** (not isolated) was monitored by TLC (R_f = 0.38). Benzyl chloride (292.1002mg, 1 equivalent) was added at this stage and the reaction mixture was stirred for another 3 hr till the intermediate compound **1b** (not isolated) was developed (monitorted by TLC; R_f = 0.40). 2 gms of solid Na_2CO_3 was added at this stage and the reaction mixture was stirred for further 1 hr while the progress of the reaction was again monitored by TLC (R_f = 0.43, 0.50). The reaction was typically completed when the N-O bond was cleaved. Basic workup, removal of pyridine hydrochloride and silica gel column chromatographic purification using ethyl acetate-hexane provided desired benzaldehyde as colourless liquid (712mg, 89% ; R_f = 0.43) and furan derivative (**2**) as pale yellow gummy liquid (88mg, 10% ; R_f = 0.50). This procedure was followed for all the substrates listed in Table 1.

Spectroscopic data for benzaldehyde (entry 1)

Yield: 712 mg (88%); colourless liquid; R_f = 0.43; IR (KBr): 1695(s), 1320(m), 770(s) cm^{-1}. ^{1}H NMR ($CDCl_3$): δ 9.80 (s, 1H, CHO), 7.30 – 7.16 (m, 5H, C_6H_5); ^{13}CNMR ($CDCl_3$): δ 198.00 (CHO), 136.20, 134.55, 132.60, 131.00 (aromatic carbons); FAB - MS (*m/z*): 106 (M^+), 105 (B.P), 77, 51, 28; HRMS-EI: Calcd. for C_6H_5CHO (M), 106.0417, Found; M^+, 106.0408.

Spectroscopic data for 2 (R=Me; α-N-methyl furan derivative; entry 1) [(E)-1-(dihydrofuran-2-(3H)-ylidene)-N-methyl methanamine)]

Yield: 88mg (10%); pale yellow gummy liquid; R_f = 0.50; IR (KBr): 3125-3054 (br), 2838 (m), 1652 (s), 1455 (m), 1210 (m) cm^{-1}; ^{1}H NMR ($CDCl_3$): δ 4.81 (br, 1H, N-H), 4.56 (s, 1H, C=CH), 3.30 (N-Me), 2.50 - 2.16 (m, 6H); ^{13}C NMR ($CDCl_3$): δ 103.00, 101.76 (double bonded carbons), 26.22, 25.30, 23.65 (3 CH_2 carbons); FAB – MS: *m/z* 113 (M^+), 98, 97; HRMS-EI: Calcd. for $C_6H_{11}ON$ (M), 113.1000, Found: M^+, 112.9876.

Spectroscopic data for propionaldehyde (entry 2)

Yield: 592mg (87%); colourless liquid; R_f = 0.50; IR (KBr): 2920 (m), 2720 (m), 1720 (s) cm^{-1}; ^{1}H NMR ($CDCl_3$): δ 9.70 (t, 1H, *J* = 6.60 Hz, -CHO), 2.30 (ddd, 2H, *J* = 6.08, 6 Hz, -CH_2), 1.00 (t, 3H, *J* = 6.30 Hz, CH_3); ^{13}CNMR ($CDCl_3$): δ 202.40 (CHO), 44.22 (CH_2 carbon), 35.55 (CH_3 carbon); FAB – MS: *m/z* 58 (M^+), 57, 29 (B.P); HRMS-EI: Calcd. for C_3H_6O (M), 58.0417, Found: M^+, 58.0403.

Spectroscopic data for 2 (R=Ph; α-N-phenyl furan derivative; entry 4) [(E)-1-(dihydrofuran-2-(3H)-ylidene)-N-phenyl methanamine)]

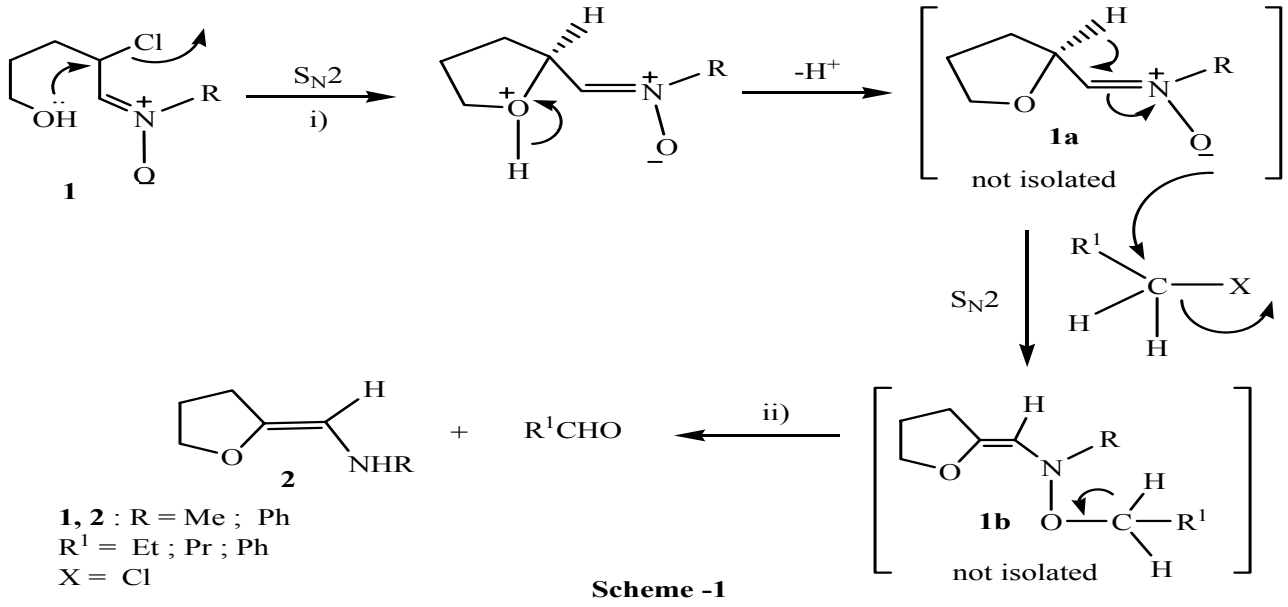
Yield: 90mg (11.5%); dark yellow viscous liquid; R_f = 0.46; IR (KBr): 3150-3060 (br), 2860 (m), 1640 (s), 1430 (m), 1140 (m), 778 (s) cm^{-1}; ^{1}H NMR ($CDCl_3$): δ 7.83 (m, 5H, C_6H_5), 6.24 (br, 1H, N-H), 2.17 (s, 1H, C=CH), 1.79 - 1.18 (m, 6H); ^{13}C NMR ($CDCl_3$): δ 137.20, 135.65, 134.00, 132.15 (aromatic carbons), 106.24, 104.18 (double bonded carbons), 28.46, 27.10, 24.84 (3 CH_2 carbons). FAB - MS (*m/z*): 175 (M^+), 98, 97, 77. HRMS-EI: Calcd. for $C_{11}H_{13}ON$ (M), 175.0993, Found; M^+, 175.0981.

Spectroscopic data for n-butyraldehyde (entry 4)

Yield: 570mg (86%); colourless liquid; R_f = 0.54; IR (KBr): 2945 (m), 2710 (m), 1730 (s) cm^{-1}; ^1H NMR ($CDCl_3$): δ 9.30 (t, 1H, *J* = 5.84 Hz, -CHO), 3.50 (dt, 2H, *J* = 6.50, 4.22 Hz, -C_2 2H), 1.30 (ddd, 2H, *J* = 5.50, 3.40 Hz, $C_3$2H), 0.90 (t, 3H, *J* = 4.30 Hz, CH_3); ^{13}CNMR ($CDCl_3$): δ 208.20 (CHO), 47.50 (C_2 carbon), 36.10 (C_3 carbon), 20.10 (C_4 carbon); FAB – MS: *m/z* 72 (M^+), 71, 57, 44 (B.P), 29; HRMS-EI: Calcd. for C_4H_8O (M), 72.0670, Found: M^+, 72.0523.

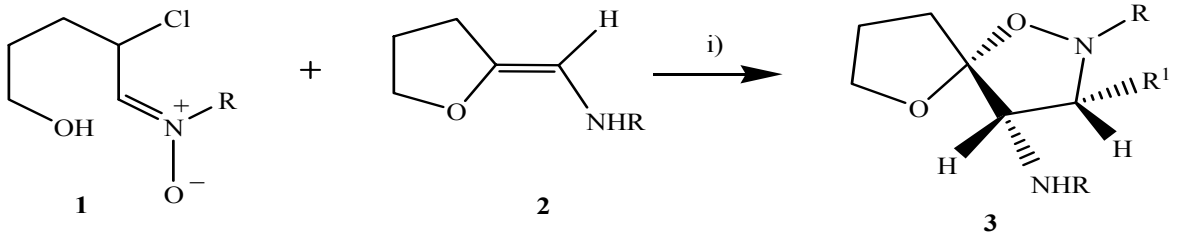
Spectroscopic data for p-hydroxy benzaldehyde (entry 5)

Yield: 776mg (89%); colourless liquid; R_f = 0.40; IR (KBr): 1690(s), 1320(m), 1210 (m), 782(s) cm^{-1}; ^1H NMR ($CDCl_3$): δ 9.76 (s, 1H, CHO), 7.05 – 6.93 (m, 4H, C_6H_5), 5.80 (s,1H, OH); ^{13}CNMR ($CDCl_3$): δ 201.64 (CHO), 134.10, 132.74, 130.40, 128.50 (aromatic carbons); FAB – MS: *m/z* 122 (M^+), 93, 92(B.P), 29; HRMS-EI: Calcd. for $C_7H_6O_2$ (M), 122.0530, Found: M^+, 122.0512.



Scheme -1

Reagents and conditions : i) Dry ether, pyridine, r.t , N_2 atmosphere
ii) Dry ether, Na_2CO_3, r.t , N_2 atmosphere



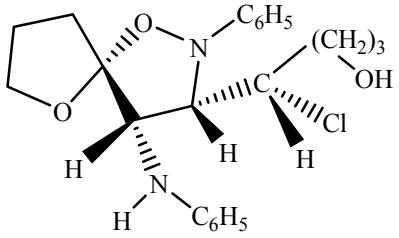
Scheme - 2

i) Reaction condition : Dry ether, RT, N_2 atmosphere, 5 - 8 hr

General procedure for cycloaddition reaction of nitrone 1 (R = Ph) with furan derivative 2 (R = Ph)

To a stirred solution of *N*–phenyl-α-chloro nitrone **1** (R = Ph; 61.8375 mg, 0.2855 mmol) in 25 mL dry ether was added **2** (R = Ph, 50 mg, 0.2855 mmol, 1 equivalent) and stirred at RT with a magnetic stirrer

under N_2 atmosphere for 5 hr. The progress of the reaction was monitored by TLC (R_f = 0.46). After completion of the reaction, the solvent was evaporated using a rotary evaporator to afford crude cycloadduct **3** (R=Ph) which was purified by column chromatography using ethyl acetate - hexane and was obtained as dark red viscous liquid **3** (R=Ph; 95 mg, 85% ; Scheme-2). This procedure was followed for the synthesis of other spiro cycloadducts **3** (R=Me).

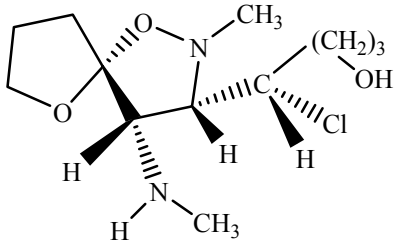


3 (R=Ph)

(S)-4-chloro-4-((3S,4S,5R)-2-phenyl-4-(phenylamino)-1,6-dioxa-2-azaspiro[4.4]nonan-3-yl)butan-1-ol

Spectroscopic data for 3 (R = Ph)

Yield: 95mg (85%); dark red viscous liquid; R_f = 0.46; IR ($CHCl_3$): 3485 – 3290 (br), 2825 (m), 2425 (m), 1620 (s), 1445 (m), 1260 (m), 1040 (m), 780 (s) cm^{-1}. ^1H NMR ($CDCl_3$): δ 6.98 - 6.93 (m, 10H, 2 x C_6H_5), 5.84 (d, 1H, *J* = 9.20 Hz, C_4H), 4.96 (br, 1H, CH_2OH, exchanged in D_2O), 3.51 (dd, 1H, *J* = 9.34, 7.88 Hz, C_3H), 3.45 (s, 1H, N – H proton), 2.61 (dt, 1H, *J* = 9.44, 8.72 Hz, CHCl), 1.88 – 1.15 (m, 12H). ^{13}C NMR ($CDCl_3$): δ 138.00, 136.50, 134.30, 133.80, 131.75, 130.42, 129.46, 128.64 (aromatic carbons), 95.10 (CHCl), 86.40 (C_5), 73.75 (C_3), 53.30 (C_4), 30.20, 28.55, 27.34, 26.22, 25.73, 24.37 (6 CH_2 carbons). MS (*m/z*): 404 (M^++2), 402 (M^+), 325, 310, 309, 218 (B.P), 107, 91, 77. HRMS-EI: Calcd. for $C_{22}H_{27}O_3N_2Cl$ (M), 402.7130, Found; M^+, 402.7122.

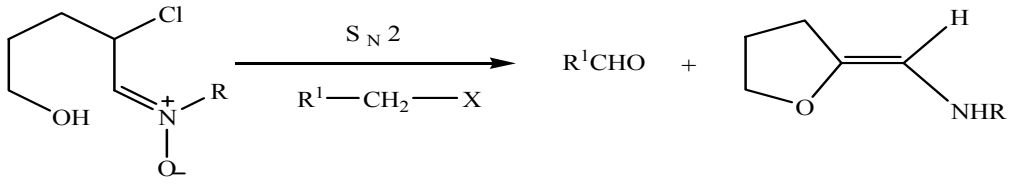


3 (R=Me)

(S)-4-chloro-4-((3S,4S,5R)-2methyl-4-(methylamino)-1,6-dioxa-2-azaspiro[4.4]nonan-3-yl)butan-1-ol

Spectroscopic data for 3 (R = Me)

Yield: 91mg (83%); red gummy liquid; R_f = 0.40; IR ($CHCl_3$): 3460 – 3326 (br), 2835 (m), 2420 (m), 1440 (m), 1325 (m), 980 (m) cm^{-1}. ^1H NMR ($CDCl_3$): δ 4.83 (br, 1H, CH_2OH, exchanged in D_2O), 4.50 (br, 1H, $NHCH_3$), 3.31 (s, 6H, 2 x N-CH_3), 2.99 (d, 1H, *J* = 9.16 Hz, C_4H), 2.50 (dd, 1H, *J* = 9.06, 7.60 Hz, C_3H), 2.19 (dt, 1H, *J* = 9.16, 8.50 Hz, CHCl), 1.66 – 1.60 (m, 12H). ^{13}C NMR ($CDCl_3$): δ 93.00 (CHCl), 87.55 (C_5), 76.20 (C_3), 55.20 (C_4), 41.97 (N-CH_3), 40.24 (NH-CH_3), 33.37, 31.50, 28.68, 26.00, 25.12, 23.40 (6 CH_2 carbons). MS (*m/z*): 280 (M^++2), 278 (M^+), 263, 248, 156 (B.P), 141, 107. HRMS-EI: Calcd. for $C_{12}H_{23}O_3N_2Cl$ (M), 278.6710, Found; M^+, 278.6698.



R = Me ; Ph R^1 = Et; Pr; Ph X = C l

Table-1 : Aldehyde synthesis using α-chloro nitrones

Yield %	Entry	Nitrone	Alkyl halide[a]	Product[b]	Time Hrs
88	1	R = Me	Benzyl chloride	Benzaldehyde	5
87	2	R = Me	1-chloro propane	Propionaldehyde	6
88	3	R = Ph	Benzyl chloride	Benzaldehyde	5
86	4	R = Ph	n-butyl chloride	n-Butyraldehyde	6
89	5	R = Me	p-hydroxy benzyl chloride	p-hydroxy benzaldehyde	4
89	6	R = Ph	p-hydroxy benzyl chloride	p-hydroxy benzaldehyde	5

[a] Reaction condition:α-chloro nitrone (3.0198 mmol),alkyl halide (1 equivalent),dry ether,Py,Na_2CO_3,N_2 atmosphere, RT

[b] All the compounds were characterized by IR, ^{1}H NMR, ^{13}C NMR, MS, HRMS spectral data.

[c] Isolated yield after purification.

RESULTS AND DISCUSSION

α-chloro nitrones (**1**) are moderately stable and can be isolated while transient nitrone **1a** can not be isolated because of its high unstability and undergoes decomposition at room temperature. The lone pair of electron of the OH group of α-chloro nitrone facilitates intramolecular S_N2 reaction in presence of pyridine and is actually the driving force for the development of transient nitrone **1a**. Nitrone **1a** reacts very quickly with different alkyl halides (S_N2 reaction) and develops an intermediate compound (**1b**). The labile N-O bond of **1b** undergoes cleavage[12] when the reaction mixture is stirred with solid sodium carbonate which plays an important role for the development of aldehyde and furan derivative (**2**) as side product in a Kornblum type process (Scheme-1;Table 1). The novelty of the study is the use of α-chloro nitrone as an oxidizing reagent in aldehyde synthesis and newly developed side product as novel dipolarophile in cycloaddition reactions. The isolated side products (**2**) are equally efficient like other conventional dipolarophiles used for cycloaddition reactions and leads to the formation of regioselective 5-substituted spiro cycloadduct (**3**)[13,14] in 1,3-dipolar cycloaddition reaction with nitrone **1** (Scheme-2) and thereby offering greater scope for its applications. The yield of the isolated aldehydes are extremely high (**85 - 89%**) in a much lesser time and are much better in case of active alkyl halides compared to inactive alkyl halides. The results are summarized in Table 1. The beauty of the reaction lies in addition of pyridine at the begining to generate transient nitrone (**1a**) which is only capable of developing furan derivative (**2**) as side product and can be utilized as a new efficient dipolarophile in 1,3-DCR and thereby the reaction as a whole becomes atom efficient. Simple nitrones[15](benzaldehyde derived nitrone) can also be employed as an oxidizing reagent for aldehyde synthesis (synthesized propionaldehyde:yield **67%**) but the side product obtained is a waste and can not be used for further reactions. At the outset of this work it was not clear about the development of transient nitrone (**1a**) but after completion of the study and spectral analysis of side product (**2**) the development of transient nitrone **1a** was confirmed. The products especially aldehydes are known compounds and spectral data of the synthesized aldehydes are almost

identical to the values found in literature. For example, sharp singlet signals at δ 9.80 & 198.00 in the NMR spectrum (^{1}H, ^{13}C respectively) along with molecular ion peak at 106, base peak at 105 and peaks at 77, 51 in the MS spectrum give strong evidence in favour of benzaldehyde formation. The oxidation side product (**2**) was obtained as single isomer having *E* configuration in all the cases and the yield of the side product was almost **10 – 13%** when isolated in pure condition.The spectral data of the oxidation side products (**2**) also agreed well with the assigned structures. The spiro cycloadducts (**3**) were obtained as regioselective single isomer predominantly in 1,3-DCR of α-chloro nitrone (**1**) with side product (**2**) having high yields (**70 – 85%**) when isolated in pure condition. The stereochemistry of the 5-substituted regioselective spiro cycloadducts (**3**) in all the cases were rationalized by considering the multiplicity of the proton signals at 3-H, 4-H and CHCl asymmetric centres along with their coupling constant values.[16] In the spiro isoxazolidine derivatives (**3**), 3-H resonates around δ_H 3.50 to 2.50 ppm while for the 4–H around δ_H 5.80 to 3.00 ppm and the coupling constant is $J_{3,4}$ ~ 9.20 Hz implying a *cis* relationship between H-3 and H-4. The CHCl proton also resonates around δ_H 2.60 to 2.20 ppm. The 3-H and CHCl protons are also *syn* as evidenced from their coupling constant values ($J_{3,CHCl}$ ~ 9.16 to 9.40 Hz).[16] Cycloaddition of *Z* nitrone (both the reported α-chloro nitrones are of *Z* configuration in this communication) via *exo* transition state geometry results in the formation of *syn* isoxazolidine derivatives. Cycloaddition reaction using furan derivatives (**2**) with other simple nitrones[15,17,18] are in progress using the same methodology. A preferential conformation for the spiro regioselective isoxazolidine derivatives (**3**) may be represented in Figure 1. Reaction of nitrone **1** with methyl iodide and ethyl bromide was also studied for the synthesis of formaldehyde and acetaldehyde respectively but no significant results were obtained because of the volatility of formaldehyde and difficulculties associated with the synthesis of acetaldehyde. These are the drawbacks of this methodology.

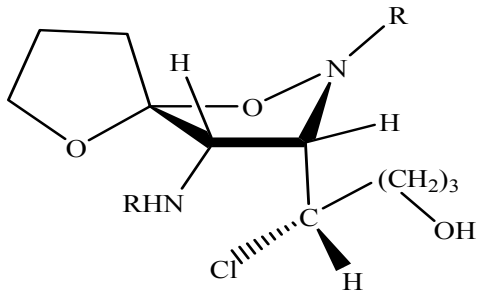


Fig 1- General conformation for the cycloadducts **3**

CONCLUSION

Finally, we developed a new atom efficient methodology for the aldehyde synthesis using α-chloro nitrone as oxidizing reagent and considered further reaction carried out on the side product with α-chloro nitrones in 1,3-dipolar cycloaddition reaction for the development of stereochemically important 5-spiro isoxazolidines. The formation of the desired cycloadducts were obtained in good yields within a short reaction time. The newly developed side products (furan derivatives, **2**) are equally effective as dipolarophile in cycloaddition reactions like other conventional dipolarophiles used for cycloaddition reactions. The notable advantages offered by this method are one pot synthesis, simple operation, easy workup, mild and faster reaction conditions with high yield of products.

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