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N-hydroxyphthalimide (NHPI) Promoted Aerobic Baeyer-Villiger Oxidation in the Presence of Aldehydes

Lingyao Wang,[a] Yongtao Wang,[a] Renfeng Du,[a] Rina Dao,[a] Haoran Yuan,[a] Cheng Liang,[b] Jia Yao,[a] and Haoran Li*[a, b]

Dedication ((optional))

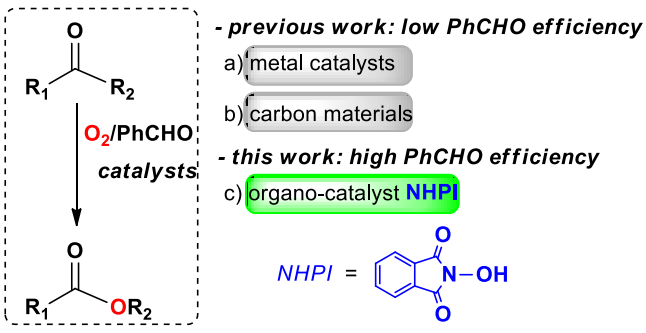
Abstract: Metal-free aerobic Baeyer-Villiger (BV) oxidation of ketones to lactones or esters in the presence of aldehydes promoted by *N*-hydroxyphthalimide (NHPI) has been developed. The reaction proceeded under mild conditions with excellent selectivity and high yields. Compared with the methods that use metal complexes as catalysts, this strategy not only showed good environmental advantages, but also increased aldehyde efficiency up to 84%. Control experiments indicated that NHPI accelerated the oxidation of aldehydes to peroxyacids but did not improve the BV oxidation while peroxyacids were already generated. Peroxyacids generated from aldehydes *in situ* were the key intermediates, and the phthalimide-*N*-oxyl radical (PINO) contributed to high aldehyde efficiency by stabilizing the radical species, which are necessary for the chain propagation reactions. This study may offer some useful strategies for new transition metal-free catalytic aerobic oxidation reactions in which aldehydes act as sacrificial agents.

Introduction

The Baeyer-Villiger (BV) oxidation is one of the most attractive reactions to convert ketones into corresponding esters or lactones directly.[1] It has inspired great interest in academia as well as industry due to the high availability to produce important intermediates in pharmaceuticals, herbicides and polymers.[2] It was first reported in 1899 for the oxidation of the menthone and tetrahydrocarvone to the corresponding lactones using Caro's acid by Baeyer and Villiger.[3] In the classical BV reaction, organic peroxyacids, such as monopersulfuric acid, perbenzoic acid, *m*-chloroperbenzoic acid (*m*-CPBA) are generally used as oxidants.[4] However, their unstability, shock sensitivity and potential explosiveness have severely curtailed the transport and storage and limited their commercial application. An environmentally benign oxidant is molecular oxygen, which is natural, inexpensive and safe.[2c, 5] Since Mukaiyama *et al.*[6] disclosed that aerobic BV oxidation can be achieved through *in situ* generation of organic peroxyacids *via* joint use of molecular oxygen and aldehydes catalyzed by Nickel (II) complexes coordinated with 1,3-diletones, increasing attention has been focused on the exploitation of other efficient catalysts.

So far, a plenty of homogeneous/heterogeneous catalysts with activity for BV oxidation have been described, such as metal oxides (*eg*: Fe_2O_3, RuO_2, MnO_2 and SnO_2),[7] metal complexes (*eg*: $Cu(OAc)_2$, $Ni(OAc)_2$, $Ni(oxa)_2$, $Co(acac)_2$, and Fe(TPP)Cl),[8] and metal based materials (*eg*: Mg-Al-Cu-CO_3 hydrotalcite, Cu-MCM-41, Fe-MCM-41-DHT, Fe-TCPP-PMO, m-Zr-P and SnTPP/4A-MS).[9] To avoid the detrimental environmental pollution caused by metal catalyst residues, it is required to develop effective metal-free catalysts. Recently, carbon materials have been reported as metal-free heterogeneous catalysts for BV oxidation.[10] In terms of homogeneous metal-free catalysts, rare examples have been reported.[11]

N-Hydroxyphthalimide (NHPI) is an effective organocatalyst for the aerobic oxidation of organic compounds *via* acting as a precursor of the phthalimide-*N*-oxyl radical (PINO).[12] Most of the examples reported in literatures described the utilization of NHPI with transition-metal complexes as co-catalysts.[13] However, the standards of green chemistry imposed the development of nonmetallic processes. Therefore, many efforts have been devoted to the design of new non-metal initiators for the activation of NHPI.[14] In 1997, Einhorn *et al.* reported an aerobic oxidation of hydrocarbons to alcohols and ketones mediated by NHPI and acetaldehyde.[15] Afterwards, Akichika and co-workers reported an epoxidation protocol of various alkenes with molecular oxygen and benzaldehyde under visible light irradiation of fluorescent lamp.[16] Nevertheless , the application of NHPI in BV oxidation has never been described. Based on the previous work,[17] we reported herein the use of NHPI (**Scheme 1**) as a catalyst for the BV oxidation of ketones with molecular oxygen and aldehydes. The reaction occurred under mild conditions with good yields and selectivity. Compared with the reported method,[9-11] this strategy not only showed good environmental advantages, but also increased aldehyde efficiency (**Table S1**).



Scheme 1. Aerobic Baeyer-Villiger (BV) oxidation of ketones

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Results and Discussion

The initial reaction of cyclohexanone with 2 equivalents of benzaldehyde in 1,2-dichloroethane in the presence of 10 mol% of NHPI at 30ºC for 4 h only gave ε-caprolactone in 3% yield (**Table 1**, entry 1). 97% of cyclohexanone and 94% of PhCHO remained unreacted due to the low reaction rate. The yield was increased to 35% when the temperature rose to 35ºC and was further improved to 96% at 40ºC (entries 2, 3). The continuous increase of temperature to 45ºC and 50ºC resulted in the decrease of yields (entries 4, 5). Meanwhile more of cyclohexanone and PhCHO were left unreacted, which was probably because of the partial decomposition of NHPI under high temperature. Other solvents such as MeCN, EtOAc and toluene were not effective (entries 6-8). The amount of NHPI was vital to the reaction. Varing the loading of NHPI to 5 mol% or 15 mol% led to the lower yields of ε-caprolactone. PhCHO as the sacrifice was also important to the reaction. Reducing the amount of PhCHO resulted in a small decrease of yields (entry 11). However, the yields was still as high as 84% even with 1 equivalent of PhCHO (entry 12), and the aldehyde efficiency was raised to 84% (Table S1), which was higher than any other reported value using metal or carbon catalysts. There was no reaction when using TEMPO instead of NHPI as the catalyst. More detailed screening conditions are listed in the SI (**Table S2-5**).

Other kinds of aldehydes were also tested and the results were listed in Table 2. Substituted benzaldehydes such as 4-methylbenzaldehyde, 4-tert-butylbenzaldehyde, 4-fluro-benzaldehyde, 4-chloro-benzaldehyde and 3-chloro-benzaldehyde were also efficient for the conversion (entry 2-6). However, *p*-anisaldehyde, 4-hydroxy-3-nitro-benzaldehyde, 4-nitro-benzaldehyde and 2-hydroxy-benzaldehyde were not effective at all (entries 7-10). Isobutyraldehyde and heptanal were less efficient but still provided ε-caprolactone in 85.9 and 81.3% yields respectively (entries 11, 12). Considering the lower commercial price of aliphatic aldehydes, it is meaningful that isobutyraldehyde and heptanal can serve as effective sacrificing agents in this oxidation system.

We then tested the scope of the reaction. In general, the reaction consistently displayed high yields with excellent selectivity under mild conditions. The yield of ε-caprolactone rose to 99.6% when the reaction time prolonged to 4.5 h (**Table 3**, entry 1). 4-, 3-, and 2-methylcyclohexanone were converted into products in 97.7%, 94.6% and 97.8% yields respectively in 8 h (entries 2-4). Other ketones like cyclopentanone, 2-norbornanone and 2-adamantanone were also oxidized effectively under the same catalytic conditions in high yields with longer reaction time (entries 5-7). The only exception was pinacolin, which has scanty conversion of 14% after 18h (entry 8). The stereochemistry and reactivity followed the tendency reported in other literature for BV oxidation.[7-11]

The BV oxidation of ketones in the presence of aldehydes usually contained two steps: 1) oxidation of aldehydes to peroxyacids, and 2) oxidation of ketones to esters by peroxylacids. To clarify the role that NHPI played in step 1, reactions in the absence of ketones were carried out under the

Table 1. Optimization of the reaction conditions.[a]

Entry	Solvent	NHPI/PhCHO[b]	T (°C)	Yield(%)[c]
1	DCE	0.1:2	30	3
2	DCE	0.1:2	35	35
3	**DCE**	**0.1:2**	**40**	**96**
4	DCE	0.1:2	45	89
5	DCE	0.1:2	50	73
6	MeCN	0.1:2	40	15
7	EtOAc	0.1:2	40	33
8	Toluene	0.1:2	40	11
9	DCE	0.05:2	40	19
10	DCE	0.15:2	40	61
11	DCE	0.1:1.5	40	93
12	DCE	0.1:1	40	84
13[d]	DCE	0.1:2	40	0

[a] Reactions were conducted on a 2.0 mmol scale in 20 mL of the solvent in a round bottom flask with an O_2 balloon for 4h. [b] The ratio is based on the substrate. [c] Yield was calculated by GC measurement based on the internal standard method (biphenyl). [d] TEMPO was used instead of NHPI.

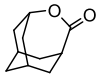
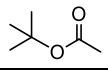
Table 2. The BV oxidation of cyclohexanone in the presence of different aldehydes. [a]

Entry	Aldehydes	Time (h)	Conv. (%) [b]	Sel. (%)[b]
1	PhCHO	4.5	100	99.6
2	4-Me-PhCHO	8	100	99.9
3	4-*t*Bu-PhCHO	24	94.7	99.9
4	4-F-PhCHO	8	100	99.9
5	4-Cl-PhCHO	12	100	99.9
6	3-Cl-PhCHO	8	100	99.9
7	4-MeO-PhCHO	24	trace	-
8	4-OH-3-NO_2-PhCHO	24	trace	-
9	4-NO_2-PhCHO	24	trace	-
10	2-OH-PhCHO	24	trace	-
11	*i*Pr-CHO	18	93.3	91.1
12	*n*-C_6H_{13}-CHO	18	81.6	99.6

[a] Reactions were conducted on a 2.0 mmol scale in 20 mL of the DCE in a round bottom flask with an O_2 balloon.
[b] Conversion and selectivity were calculated by GC measurement based on the internal standard method (biphenyl); selectivity = yield/conversion.

same conditions as the catalytic tests. The conversion of PhCHO was analyzed by GC. The result showed that the PhCHO was completely consumed in 4 h in the presence of NHPI. However, the conversion of PhCHO was less than 3% in 4 h without NHPI. The autoxidation of PhCHO without solvents

Table 3. BV oxidation of various ketones. [a]

Entry	Substrates	Products	*T.* (h)	Conv./Sel./Yield. (%) [b]		
1			4.5	100	99.6	99.6
2			8	97.8	99.9	97.7
3			8	100	94.6	94.6
4[c]			8	97.9	99.9	97.8
5			18	94.2	95.5	90.0
6			18	92.0	99.9	91.9
7			18	91.0	99.9	90.9
8			18	14.0	99.3	13.9

[a] Reactions were conducted on a 2.0 mmol scale in 20 mL of the DCE in a round bottom flask with an O_2 balloon. [b] Conversion, selectivity and yields were calculated by GC measurement based on the internal standard method (biphenyl). [c] GC showed an overlapping lactone peak which was attributed to a mixture of 5-methyloxepan-2-one and 7-methyloxepan-2-one indicated by GC-MS. The selectivity and yields were for both isomers.

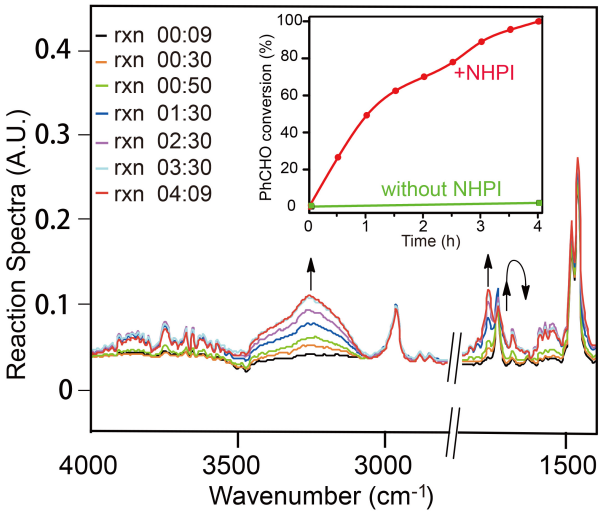


Figure 1. *In-situ* IR spectroscopy and time curve of the benzaldehyde oxidation catalyzed by NHPI.

was faster due to the increase of concentration and gave 84% conversion in 4 h (**Table S6**, entry 2). This result was consistent with the reported literature.[18] The reaction was also studied by *in-situ* infrared (IR) spectroscopy (**Figure 1**). The unchanged absorption of ~2960 cm^{-1} was from the vibration of C-H bonds in 1,2-dichloroethane (solvent). The stretching vibration absorption of C=O bond of benzaldehyde was at ~1700 cm^{-1}. The OH and C=O stretching vibration absorption of the catalyst NHPI was inconspicuous probably due to low concentration. As the reaction went by, two new signals at ~3260 cm^{-1} and ~1740 cm^{-1} appeared and became stronger with time going on, which were mainly from benzoic acid. The strength of the peak at ~1700 cm^{-1} increased first and then decreased probably due to the cooperative effects from benzaldehyde, perbenzoic acid and benzoic acid. These indicated that NHPI as a precursor of PINO radical efficiently catalyzed the oxidation of benzaldehyde.

To distinguish whether NHPI has influence on step 2, reactions with sole *m*-CPBA and *m*-CPBA/NHPI were conducted respectively (**Table 4**). The combination of NHPI and *m*-CPBA resulted in a slight decrease of the conversion when compared to the single use of *m*-CPBA probably due to the consumption of *m*-CPBA by side reaction with NHPI in higher concentration. Such experiments demonstrated that NHPI did not have positive effect on the step of oxidation of ketones to esters.

Table 4. Cyclohexanone oxidation by *m*-CPBA.[a]

Entry	Catalyst	Conv. (%)	Sel. (%)	Yield (%)
1	/	84	96	80
2	NHPI	78	96	75

a) General conditions: cyclohexanone, 2.0 mmol; *m*-CPBA, 2.0 mmol; NHPI, 10 mol%; 1,2-dichloroethane, 20 mL; O_2; 40 °C; 4 h.

EPR experiments were carried out to detect the radicals involved in the reaction. As benzoylperoxyl radical can oxidize NHPI to PINO,[15] there is a strong radical signal from PINO (A = 4.72 G, g = 2.0069) when combining PhCHO and NHPI (**Figure 2**, black curve). After the addition of cyclohexanone to the solution, the PINO signal was immediately attenuated (red curve). After reacting for half an hour, the PINO signal dropped to a very weak level (blue curve). These indicated that cyclohexanone consumed benzoylperoxyl radical and thus reduced the generation of PINO radical. No EPR signals were observed in the blank experiments of $PhCHO/O_2$ or $NHPI/O_2$ system.

The features of N-oxyl radicals as catalysts are crucially dependent on the O-H bond dissociation energy (i.e. hemolytic dissociation enthalpies (BDEs) of the O-H bond) of the radical precursors.[19] The BDE of TEMPOH and NHPI is 69.6 and 83.5 kcal/mol respectively, while that of PhCHO is 89.6 kcal/mol.[19a, 20] Owing to the low BDE of TEMPOH, the thermodynamic driving force for direct oxidation *via* H-abstraction by TEMPO is low in most circumstances. Thus C-H activation using this route can hardly be complied. On the contrary, the BDE of NHPI is significantly larger and hence the corresponding PINO radical is a much stronger H-abstracting reagent. PINO played a key role in propagating free-radical reaction chains, while TEMPO inhibited free-radical processes. To investigate the nature of the

induction period, we conducted a series of experiments using TEMPO as a free radical inhibitor (**Figure 3**). After the cyclohexanone oxidation reacted under the general condition for an hour, 10 mol% of TEMPO was added to the system. The EPR signal of TEMPO (A = 15.82 G, g = 2.0055) gradually diminished until it disappeared. Analysis of the reaction with TEMPO addition by GC-MS displayed a signal with *m/z* matching the expected mass of the TEMPOOCPh adduct formed from the capture of the benzaldehyde radical by TEMPO (**Scheme 2**). These experiments suggested that TEMPO terminated the radical chain process by coupling with the benzoyl radical.

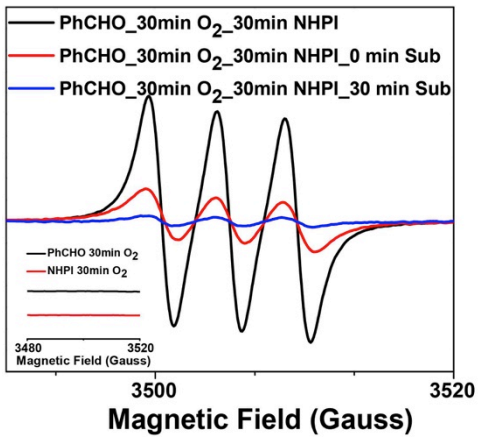


Figure 2. EPR spectra of the mixture of NHPI and PhCHO (black) with the addition of cyclohexanone immediately (red), for 30 minutes (blue). Control experiments of PhCHO/O_2 (black, left down) and NHPI/O_2 (red, left down).

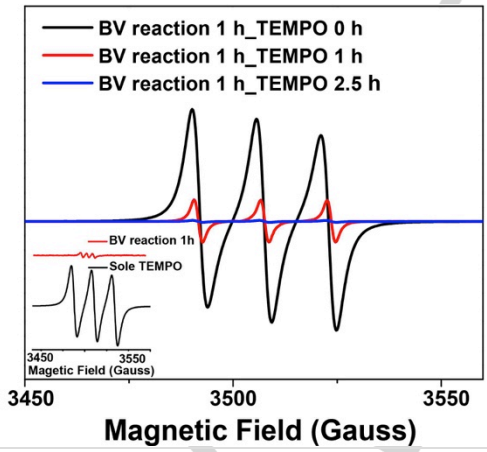


Figure 3. EPR spectra of the typical reaction after the addition of TEMPO at t=0 h (black), t=1 h (red), t=2.5 h (blue). Typical experiments of BV oxidation for 1 h without TEMPO (red, left down) and sole TEMPO (black, left down).

More TEMPO addition experiments were conducted and the results were shown in **Figure 4**. The addition of 10 mol% of TEMPO at the beginning of the reaction (pink) virtually delayed the product formation in contrast to the reaction without TEMPO (black). During the initiation period (*i.e.*, at 1 h with 30% conversion, red and blue), 10 mol% and 100 mol% of TEMPO were added to the reaction system respectively. The former continued to react with a lower conversion rate, reaching to a conversion of 66%; and the latter ceased immediately (**Figure 4a**). Similar TEMPO addition reactions using one equivalent of *m*-CPBA as the direct oxidant were also performed. The reaction rate was faster but the yield and selectivity were slightly inferior to that of PhCHO-NHPI-O_2 system (black). The addition of 10 mol% (red) of TEMPO and 100 mol% (blue) of TEMPO reduced the oxidation rate and decreased the conversion of cyclohexanone by about 18% and 51% respectively. Adding 100 mol% TEMPO into the mixture after 20 min, the reaction slowed down (pink) (**Figure 4b**). These experiments suggested the similarity of PhCHO-NHPI-O_2 and *m*-CPBA systems and the perbenzoic acid generated *in-situ* in the PhCHO-NHPI-O_2 system was the final oxidant for BV oxidation.

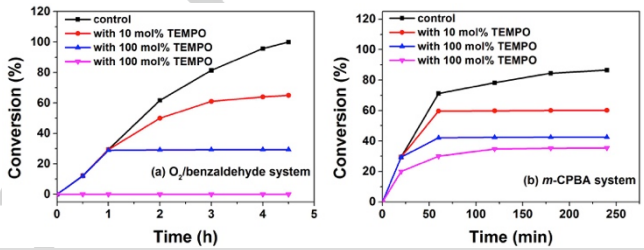
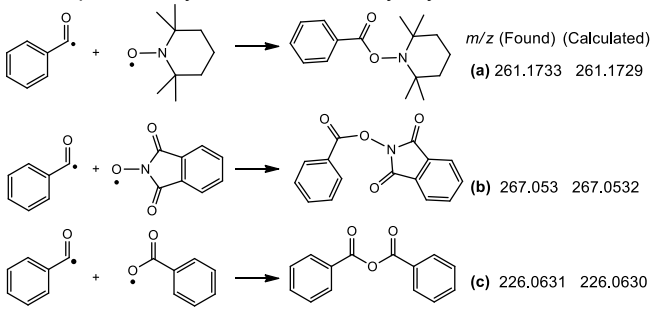


Figure 4. Reaction profiles with TEMPO addition. (a) Reaction conditions: cyclohexanone, 2.0 mmol; benzaldehyde, 4.0 mmol; NHPI, 10 mol%; 1,2-dichloroethane, 20 mL; O2; 40°C (black). With 10 mol% of TEMPO added during the reaction at t = 1 h (red). With 100 mol% of TEMPO added during the reaction at t = 1 h (blue). With 10 mol% of TEMPO added at the beginning of the reaction (pink). (b) Reaction conditions: cyclohexanone, 2.0 mmol; *m*-CPBA, 2.0 mmol; 1, 2-dichloroethane, 20 mL (black). With 10 mol% of TEMPO added during the reaction at t = 20 min (red). With 100 mol% of TEMPO added during the reaction at t = 20 min (blue). With 100 mol% of TEMPO added at the beginning of the reaction (pink).

To get more hints of the radical intermediates of the reaction, we increased the amount of NHPI to one equivalent to improve the radical concentrations and used GC-MS to capture the possible intermediate adducts. Except for the TEMPOOCPh adduct **a**, two new radical adducts were observed (**Scheme 2**). The adduct **b** consisted of benzoyl radical and PINO while **c** was made up of benzoyl radical and benzoyloxy radical.



Scheme 2. The capture of intermediate radical adducts by GC-MS.

Based on the above control experiments, a possible mechanism for this aldehyde-NHPI-O_2 oxidation system was proposed (**Figure 5**). At first, the reaction was initiated by the slow autoxidation of aldehydes with O_2 (pathway A) to form acyl radicals (I), which were very reactive to produce acylperoxyl radical intermediates (II) by O_2 insertion. The radicals (II)

abstracted hydrogen from NHPI to form peroxyacid (III) and PINO. PINO may in turn oxidize aldehydes to radicals (I). The oxidation of aldehydes by PINO H-abstraction was much faster than the initial autoxidation. Once a sufficient amount of radicals existed in the system, the chain propagation reaction would be accelerated. Peroxyacids formed *in situ* attacked the carbonyl group of ketones to generate the "Criegee adduct intermediates", which would undergo rearrangement to produce lactones/esters and carboxylic acid.[21]

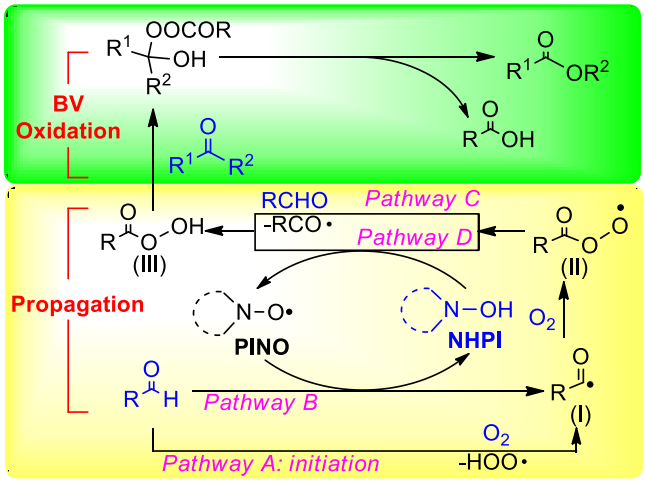


Figure 5. Proposed mechanism

Conclusions

In conclusion, we disclosed a NHPI mediated aerobic BV oxidation of ketones in the presence of aldehydes. The reactions proceeded under mild conditions and displayed excellent selectivity and high yields. NHPI promoted the transformation by accelerating the oxidation of aldehydes to peroxyacids, which were the key intermediates. PINO generated from NHPI contributed to higher aldehyde efficiency by stabilizing radical species that are necessary for the chain propagation reactions. This aldehyde-NHPI-O_2 system represented a novel metal-free approach for efficient BV oxidation that exhibited better environmental and economical advantages. Our study may offer some useful strategies for green catalytic aerobic oxidation reactions systems in which aldehydes act as sacrificial agents. More efforts should be done to clarify the detailed reaction mechanism.

Experimental Section

Materials. Cyclohexanone (>99.0%), *tert*-butyl acetate (>99.0%), 2-norbornanone (>98%), biphenyl (99.5%), 2-methylcyclohexanone (>96.0%), 3-methylcyclohexanone (>97.0%), were purchased from TCI without further purification unless indicated. 2-adamantanone (99%), 2,2,6,6-tetramethyl-1-piperidinyloxy (TEMPO) (98%), *N*-hydroxyphthalimide (NHPI) (98%), benzaldehyde (98%), were purchased from J & K Scientific Ltd. without further purification unless indicated. Acetonitrile, toluene, ethyl acetate were analytical grade which were purchased from J & K Scientific Ltd. 1,2-Dichloroethane (DCE) (99.5%), 3-chloroperoxybenzoic acid (85%), 3-chlorobenzaldehyde (98%), cyclopentanone (99%), heptanal (97%) were purchased from Energy Chemical without further purification unless indicated. Methyl trimethylacetate (99%), isobutylaldehyde (98%) were purchased from Tansoole without further purification. Other chemicals were purchased from Adamas-beta without further purification.

General Methods. The progress of the reaction was monitored by taking samples at various intervals to be analyzed using gas chromatograph (GC) (Agilent 7820A) equipped with a DB-35/ZB-35/HP-35 column (30 m × 0.32 mm × 0.25 mm) and a Flame Ionization Detector (FID). The conversion was calculated on the basis of the peak area ratio of ketones against the internal standard, biphenyl. The product yields were calculated on the basis of the peak area ratio of lactones or esters against the internal standard. The structural analysis of target product was conducted on a Gas chromatography/ Mass spectrometry (GC/MS) (Agilent 7200-Q-TOF). EPR spectra were obtained by using a computer controlled X-band (9.5GHz) EPR spectrometer (Bruker A300). *In-situ* IR Spectrum was measured by ReactIR 10 with MCT detector.

General Procedure for BV Oxidation. In a typical reaction, ketones (2 mmol), benzaldehyde (4 mmol), NHPI (10 mol%) and 1,2-dichloroethane (20 mL) were placed into a three-necked round bottom flask (50 mL) equipped with an oxygen balloon and a magnetic stir bar. The mixture was stirred at 40ºC for 4-18 hours. The reaction was monitored by GC. The conversion was calculated on the basis of the peak area ratio of ketones against the internal standard, biphenyl. The product yields were calculated on the basis of the peak area ratio of lactones or esters against the internal standard.

***In-situ* IR Spectroscopic experiments.** To a 500 mL glass flask fitted with a magnetic stir bar, an air inlet pipe, a water cooled reflux condenser was added 2.14 g (0.02 mol) of benzaldehyde, 0.16 g (0.001 mol) of NHPI and 100 mL of 1,2-dichloroethane (DCE). The mixture was stirring at 300 rpm while the temperature was held at 40ºC and air was fed at a rate of 20 ml/min. Oxygen uptake proceeded smoothly. The reaction lasted for 4 h. The reaction mixture was analyzed by an *in-situ* IR instrument (ReactIR 10 with MCT detector). The spectra were shown in Figure S1.

General EPR experiments (a) with NHPI To a 50 mL three-necked glass flask fitted with a water cooled reflux condenser, a magnetic stir bar and an oxygen balloon was added 0.4245 g (4 mmol) of benzaldehyde and 20 mL of 1,2-dichloroethane (DCE). The mixture was stirred at 40ºC for 30 min at atmospheric pressure. Then 0.0326 g (0.2 mmol) of NHPI was added into the mixture and the mixture was stirred for another 30 min. The EPR spectra of the reaction solution (Figure 2, black) were obtained by using a computer controlled X-band (9.5GHz) EPR spectrometer (Bruker A300). Thereafter, EPR was measured immediately upon addition of 0.1962 g (2 mmol) of cyclohexanone into the reaction system (red). The reaction was continued for 30 min before measuring EPR (blue). **(b) with TEMPO** To a 50 mL three-necked glass flask fitted with a water cooled reflux condenser, a magnetic stir bar and an oxygen balloon was added 0.0326 g (0.2 mmol) of NHPI, 0.4245 g (4

mmol) of benzaldehyde, 0.1962 g (2 mmol) of cyclohexanone and 20 mL of 1,2-dichloroethane (DCE). The mixture was stirred at 40ºC at atmospheric pressure. After the typical BV oxidation reacted under the general condition for an hour, 10 mol% of TEMPO was added to the system. EPR spectrum were obtained by the X-band (9.5 GHz) EPR spectrometer (Bruker A300) immediately (Figure 3, black), at t=1 h (red) and t=2.5 h (blue).

Acknowledgements ((optional))

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Keywords: BV oxidation • NHPI • metal-free • molecular oxygen • mechanistic study

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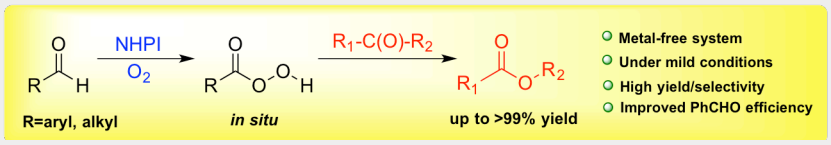

We disclosed a **metal-free** *N*-hydroxyphthalimide (NHPI) promoted aerobic Baeyer-Villiger oxidation of ketones to corresponding esters or lactones in the presence of aldehyde. The reaction proceeded **under mild conditions** with **excellent selectivity, high yields** (96%) and **good aldehyde efficiency**

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_N_-hydroxyphthalimide (NHPI) Promoted Aerobic Baeyer-Villiger Oxidation in the Presence of Aldehydes