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Iranian Chemical Communication

Original Research Article

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A facile and environmental friendly method for C=N bond cleavage of imines using *p*-toluenesulfonic acid in solid state

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Received: 13 February 2016, Accepted: 15 June 2016, Published: 15 June 2016

Abstract

A simple, efficient and clean procedure has been developed for the cleavage of imines C=N bond. Deprotection of imines to their parent carbonyl and amine compounds was achieved using *p*-toluenesulfonic acid in the solid state condition at 25-45 ˚C. The salient features of this methodology are shorter reaction times, cheap processing, high yields of product and easy availability of the catalyst. These features make this method an attractive alternative to existing routes for deprotection of imines.

Keywords: Solid state; C=N Bond cleavage; *P*-Toluenesulfonic acid; imines.

Introduction

In the synthetic chemistry, several protecting agents have been developed for amino and carbonyl groups. A routinely used method for the protection of these functional groups involves conversion to the corresponding imines [1]. Deprotection of imine groups back to amine and carbonyl agents is a necessary step for practical use of these compounds [2]. In addition, since many valuable reactions have been developed to prepare of imines from non-carbonyl and non-amino compounds [3,4], the regeneration of these molecules from imines represents a potential way for the synthesis of amines, aldehydes and ketones. Various methods have been reported for the cleavage of imines, include the using acetic anhydride in surfactant media [5], boronic acid [6], hydrochloric acid [7,8], methoxy amine [9], metallic reagents [10-13], and surfactant/I_2/water [14]. However some of these methods suffer from major drawbacks, for example use of: toxic transition metals, non-common and not cost-effective chemicals, therefore difficult to be applied for scale up operation.

Our continued interest in the development of environmental benign methodology [15-20] prompted us to investigate a novel and simple method for the deprotection of imines utilizing inexpensive and commercially available reagent. In this paper it was disclosed that *p*-toluenesulfonic acid (*p*-TsOH) is an efficient, cheaper and easy to handle compound for the cleavage of imine C=N bond. In this protocol a wide range of imines can be deprotected to the corresponding parent carbonyl compounds in the solid state conditions (Scheme 1).

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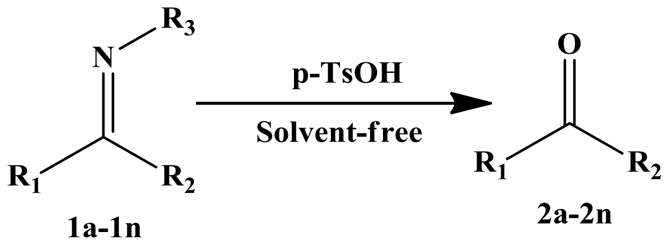


Scheme 1. Cleavage of C=N bond of imine by p-TsOH in solid state

Experimental

General

All imines were synthesized in our laboratory according to the literature procedures [24]. The progress of the reactions was followed by TLC using silica gel SILIG/UV 254 plates. ^{1}H NMR (400 MHz) and ^{13}C NMR (100 MHz) spectra were recorded on a Bruker 300 MHz instrument. FT-IR spectra were recorded on a Perkin-Elmer RX-1 instrument. The melting points were determined in open capillaries with a Stuart Melting Point Apparatus and are uncorrected.

A representative procedure for C=N bond cleavage of imines using p-toluenesulfonic acid in solid state

A well ground mixture of imine **1a** (1 mmol) and *p*-TsOH (1 mmol) was placed in a flask. This mixture was kept for the stipulated time (Table 2) at room temperature. The progress of the reaction was monitored by TLC. After completion of reaction, water (20 mL) was added to the mixture and the carbonyl product **2a** (1H-indole-2,3-dione) was extracted with Et_2O (3×5 mL). The combined organic layers were dried over $MgSO_4$, filtered, and Et_2O was evaporated under reduced pressure to obtain the pure compound **2a**.

Physical and spectroscopic data

(**1a**): mp 273-280 °C (273-278 °C). IR ν_{max}(KBr)/cm^{-1}: 511 (w), 559 (w), 634 (m), 753 (m), 845 (m), 907 (m), 992 (w), 1096 (m), 1161 (s), 1210 (m), 1284 (w), 1329 (s), 1401 (w), 1466 (s), 1536 (m), 1618 (s), 1661 (s), 3118 (w), 3292 (s). ^{1}H-NMR (DMSO, 500 MHz): δ = 2.50 (2H, s), 7.09 (1H, t, J=7.25 Hz), 7.18 (1H, t, J= 7.75 Hz), 7.31-7.38 (3H, m), 7.73 (1H, d, J=8.5), 7.75-7.91 (2H, m), 11.03 (1H, s). ^{13}C-NMR (DMSO, 125 MHz): δ = 112.04, 115.53, 117.60, 125.44, 127.47, 134.97, 140.39, 146.16, 147.29, 152.55, 155.27, 163.16. Ms m/z (%): 301 (M$^+$, 24.40), 273 (48.80), 193 (14.70), 172 (96.00), 156 (88.90), 140 (7.80), 108 (64.51), 92 (95.20), 65 (100.00).

(**1b**): mp 230-232 °C. IR ν_{max}(KBr)/cm^{-1}: 542 (w), 679 (w), 753 (w), 908 (m), 1165 (s), 1297 (w), 1339 (s), 1381 (m), 1468 (s), 1586 (m), 1608 (s), 1650 (s), 2852 (w), 2925 (m), 3103 (w), 3299 (m). ^{1}H-NMR (DMSO, 500 MHz): δ = 2.50 (2H, s), 5.02 (2H, s), 7.22-7.42 (9H, m), 7.69 (1H, t, J=7), 7.71 (1H, d, J=10), 7.90-7.93 (2H, m). ^{13}C-NMR (DMSO, 125 MHz): δ = 42.90, 110.24, 110.87, 122.48, 125.28, 126.22, 127.37, 127.47, 127.57, 128.70, 134.54, 134.72, 135.74, 139.67, 153.09, 154.30, 162.07. Ms m/z (%): 391 (M$^+$, 100.00), 363 (19.60), 300 (24.01), 236 (31.40), 220 (17.30), 207 (59.80), 172 (40.10), 156 (37.70), 108 (24.40), 91 (79.50), 65 (54.30).

(**1c**): mp 262-265 °C (260-266 °C). IR ν_{max}(KBr)/cm^{-1}: 409 (w), 434 (w), 485 (w), 520 (w), 598 (w), 633 (w), 678 (m), 714 (w), 751 (m), 797 (w), 832 (m), 880 (w), 1011 (w), 1034 (m), 1147 (w), 1197 (m), 1258 (w), 1334 (s), 1398 (w), 1463 (s), 1480 (w), 1612 (s), 1652 (w), 1722 (m), 1740 (s), 1799 (w), 2852 (w), 2924 (w), 3266 (m). ^{1}H-NMR (CDCl$_3$, 400 MHz): δ = 6.60-6.70 (2H, m), 6.80-6.93 (3H, m), 7.24 (1H, d, J=6.4 Hz), 7.34-7.36 (2H, m), 10.24 (1H, s).

(**1d**): mp 226-229 ^{o}C (225-230 ^{o}C). IR ν_{max}(KBr)/cm^{-1}: 467 (w), 510 (w), 577 (w), 630 (w), 697 (m), 735 (w), 749 (w), 776 (w), 822 (w) 850 (w), 994 (w), 1017 (w), 1096 (w), 1147 (w), 1266 (w), 1199 (m), 1236 (w), 1286 (w), 1334 (s), 1376 (w), 1403 (w), 1463 (s), 1502 (w), 1594 (w), 1612 (s), 1654 (m), 1724 (w), 1742 (s), 2919 (w), 3227 (m). ^{1}H-NMR (CDCl$_3$, 400 MHz): δ = 2.41 (3H, s), 6.81 (1H, t, J=7.6 Hz), 6.84 (1H, d, J=7.2 Hz), 6.97 (1H, d, J=7.6 Hz), 7.01 (2H, d, J=8.0 Hz), 7.28 (2H, d, J=8.0 Hz), 7.34 (1H, td, J=7.6 Hz, J=1.2 Hz), 9.41 (1H, s).

(**1e**): mp 182-188 ^{o}C (185-188 ^{o}C). IR ν_{max}(KBr)/cm^{-1}: 524 (w), 554 (m), 584 (m), 630 (w), 658 (m), 687 (w), 714 (m), 748 (w), 839 (w), 855 (s), 892 (m), 983 (w), 1012 (w), 1109 (w), 1146 (s), 1197 (w), 1304 (s), 1344 (s), 1406 (w), 1516 (s), 1585 (m), 1600 (m), 1626 (m), 1919 (w), 1445 (w), 3102 (w), 3268 (w), 3351 (s).

(**1f**): IR ν_{max}(KBr)/cm^{-1}: 517 (w), 831 (s), 869 (m), 1165 (s), 1297 (w), 1339 (s)1487 (m), 1578 (m), 1615 (m), 1887 (w), 2925 (w), 3003 (w), 3299 (s).

(**1g**): mp 117-124 ^{o}C. IR ν_{max}(KBr)/cm^{-1}: 517 (w), 533 (w), 563 (w), 679 (w), 736 (w), 806 (w), 831 (s), 869 (m), 1008 (w), 1059 (w), 1091 (s), 1137 (w), 1190 (w), 1261 (w), 1366 (w), 1384 (w), 1487 (s), 1578 (m), 1615 (m), 2925 (w).

(**1h**): mp 90-97 ^{o}C. IR ν_{max}(KBr)/cm^{-1}: 515 (w), 524 (w), 565 (w), 715 (w), 736 (w), 785 (w), 813 (s), 841 (m), 859 (w), 880 (w), 952 (w), 1015 (w), 1043 (m), 1094 (s), 1123 (w), 1184 (w), 1216 (w), 1258 (w), 1374 (m), 1466 (s), 1505 (s), 1578 (s), 1618 (m), 1899 (w), 2921 (w), 3025 (w).

(**1i**): mp 198-210 ^{o}C. IR ν_{max}(KBr)/cm^{-1}: 486 (w), 530 (m), 658 (w), 696 (m), 710 (m), 822 (s), 858 (w), 890 (m), 1067 (w), 1105 (m), 1167 (s), 1335 (s), 1434 (m), 1532 (s), 1604 (m), 2827 (w), 2912 (w), 3098 (w).

(**1j**): mp 124-128 ^{o}C (125-127 ^{o}C). IR ν_{max}(KBr)/cm^{-1}: 505 (w), 529 (m), 552 (w), 541 (w), 686 (w), 713 (w), 749 (w), 805 (m), 854 (m). 888 (w), 1009 (w), 1091 (w), 1103 (w), 1188 (w), 1342 (s), 1488 (m), 1516 (s), 1598 (m), 1624 (w), 2359 (w).

(**1k**): IR ν_{max}(KBr)/cm^{-1}: 517 (m), 571 (w), 671 (m), 691 (s), 762 (s), 845 (s), 1008 (w), 1025 (w), 1089(m), 1101 (w), 1213 (s), 1288 (m), 1313 (w), 1366 (m), 1447 (m), 1481 (s), 1627 (s), 1890 (w), 1957 (w), 2362 (w), 2927 (w), 3056 (w).

(**1l**): IR ν_{max}(KBr)/cm^{-1}: 429 (w), 588 (s), 690 (s), 760 (s), 927 (w), 955 (m), 1024 (m), 1078 (w), 1180 (w), 1266 (s), 1302 (w), 1359 (m), 1449 (m), 1582 (w), 1599 (m), 1627 (s), 1904 (w), 3005 (w), 3063 (w).

(**1m**): mp 219-223 ^{o}C. IR ν_{max}(KBr)/cm^{-1}: 460 (w), 564 (w), 579 (m), 659 (m), 693 (s), 732 (m), 767 (m), 819 (m), 853 (m), 881 (w), 966 (s), 1001 (w), 1109 (s), 1151 (m), 1178 (w), 1211 (s), 1253 (m), 1302 (s), 1317 (w), 1394 (m), 1447 (m), 1469 (s), 1575 (w), 1615 (s), 1894 (w), 3057 (w).

(**1n**): mp 127-133 ^{o}C. IR ν_{max}(KBr)/cm^{-1}: 562 (w), 648 (m), 742 (s), 774 (w), 858 (s), 899 (w), 973 (m), 1021 (s), 1024 (s), 1114 (m), 1150 (m), 1200 (m), 1219 (w), 1249 (w), 1284 (s), 1372 (w), 1419 (m), 1461 (m), 1498 (s), 1526 (w), 1578 (s), 1610 (w), 1637 (s), 1799 (w), 1943 (w), 2557 (w), 2729 (w), 2796 (w), 2931 (w).

(**2a**): mp 198-200 ^{o}C (196-202 ^{o}C). IR ν_{max}(KBr)/cm^{-1}: 456 (w), 479 (w), 637 (w), 661 (m), 736 (w), 771 (s), 818 (w), 885 (w), 946 (m), 1095 (w), 1113 (w), 1144 (w), 1189 (m), 1202 (w), 1291 (w), 1332 (s), 1403 (w), 1461 (m), 1616 (s), 1728 (s), 1897 (w), 1944 (w), 2814 (w), 2889 (w), 3060 (w), 3111 (w), 3193 (s), 3451 (w).

(**2b**): mp 128-130 ^{o}C (130-131 ^{o}C). IR ν_{max}(KBr)/cm^{-1}: 471 (m), 547 (w), 627 (m), 694 (m), 705 (w), 720 (w), 754 (m), 765 (w), 818 (w), 855 (w), 868 (w), 957 (w), 1005 (w), 1026 (w), 1076 (w), 1091 (w), 1110 (w), 1176 (m), 1198 (w), 1309 (w), 1349 (s), 1375 (w), 1442 (w), 1471 (m), 1495 (w), 1613 (s), 1732 (s), 2340 (w), 2360 (s), 3031 (w), 3451 (m).

(**2e**): mp 100-103 ^{o}C (104-107 ^{o}C). IR ν_{max}(KBr)/cm^{-1}: 412 (w), 465 (w), 512 (w), 538 (w), 678 (m), 739 (s), 817 (s), 852 (s), 1007 (w), 1104 (m), 1197 (s), 1287 (w), 1346 (s), 1539 (s), 1607 (m), 1706 (s), 1951 (w), 2851 (w), 2926 (w), 3108 (w).

(**2f**): mp 64-70 ^{o}C (68-71 ^{o}C). IR ν_{max}(KBr)/cm^{-1}: 409 (w), 419 (w), 439 (w), 553 (w), 573 (w), 662 (w), 693 (w), 759 (m), 823 (s), 847 (s), 862 (s), 1001 (w), 1048 (m), 1100 (s), 1131 (m), 1199 (s), 1250 (m), 1287 (m), 1376 (s), 1410 (w), 1460 (m), 1557 (s), 1587 (s), 1683 (s), 1788 (w), 1933 (w), 2993 (w), 3022 (w), 3088 (m).

(**2i**): mp 69-72 ^{o}C (70-74 ^{o}C). IR ν_{max}(KBr)/cm^{-1}: 508 (w), 595 (m), 632 (w), 728 (s), 813 (s), 825 (s), 938 (w), 1065 (w), 1164 (s), 1232 (s), 1314 (w), 1371 (s), 1432 (w), 1549 (m), 1594 (s), 1661 (s), 2715 (w), 2796 (w), 2912 (w).

(**2k**): bp 200 ^{o}C. IR ν_{max}(KBr)/cm^{-1}: 429 (w), 588 (s), 690 (s), 760 (s), 927 (w), 955 (m), 1024 (m), 1078 (w), 1180 (w), 1266 (s), 1302 (w), 1359 (m), 1449 (m), 1582 (w), 1599 (m), 1683 (s), 1904 (w), 3005 (w), 3063 (w).

(**2m**): mp 95-99 ^{o}C (96-98 ^{o}C). IR ν_{max}(KBr)/cm^{-1}: 440 (w), 521 (m), 647 (w), 665 (m), 703 (s), 748 (w), 764 (m), 802 (w), 825 (m), 948 (m), 1152 (m), 1183 (w), 1239 (s), 1296 (w), 1320 (s), 1464 (w), 1536 (s), 1617 (s), 3316 (s), 3420 (s).

(**2n**): bp 197 ^{o}C. IR ν_{max}(KBr)/cm^{-1}: 686 (s), 718 (s), 758 (s), 862 (w), 883 (s), 1029 (m), 1114 (m), 1151 (s), 1227 (s), 1275 (s), 1321 (w), 1386 (m), 1460 (s), 1594 (s), 1668 (s), 2751 (m), 2848 (m), 3062 (m), 3185 (m).

Results and discussion

Firstly, the deimination of **1a** was used as the model reaction in different solvents at room temperature. The best result was obtained in solvent-free condition and the product was achieved in a high yield with a shorter reaction time (Table 1, Entry 5). It is notable that both *p*-TsOH and substrate **1a** are crystalline compounds; therefore, the reaction occurred in solid state condition that is very important from environmentally point of view.

Table 1. Effect of various solvents (5 mL) on deprotection of imine **1a** with p-TsOH at r.t[a]

Entry	**Solvent**	**Time (min)**	**Yield (%)[b]**
1	Chloroform	30	-
2	Methanol	30	20
3	DMF	30	20
4	Toluene	30	-
5	non-Solvent	5	90
6	n-Hexane	30	-

[a] All reactions were run with **1a** (1 mmol) and *p*-TsOH (1 mmol) at r.t.
[b] Isolated yields

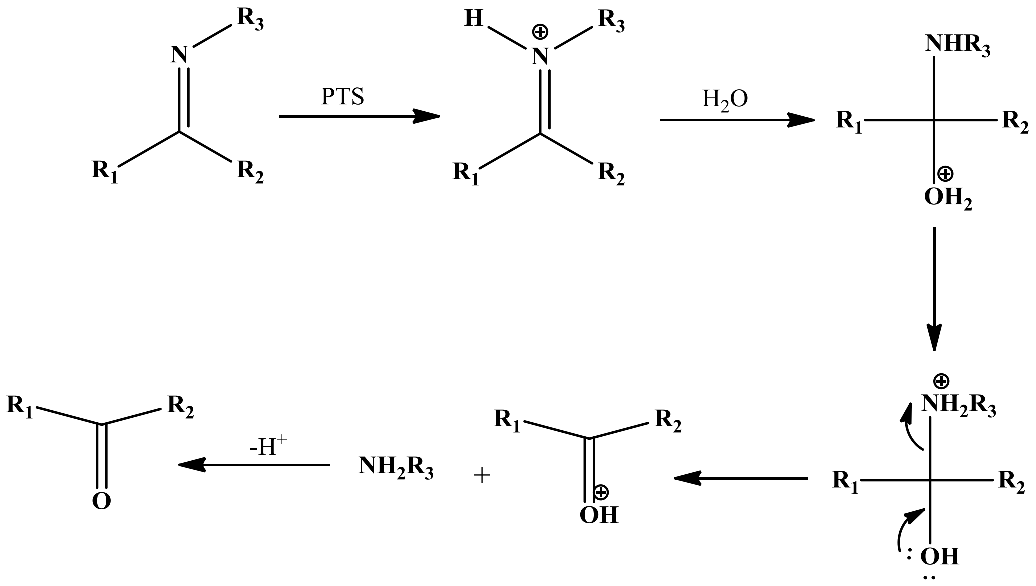
The effect of amount of *p*-TsOH on the reaction yield was also investigated on model reaction in the solid state (Table 2). Preliminary studies were conducted on 0.1 mmol amount of *p*-TsOH and 1 mmol of substrate **1a** that, within 4 h, a trace of product was examined by TLC at room temperature. Whereas, when the amount of *p*-TsOH was 0.5 mmol, the carbonyl product was obtained in 50% yield within the same time (Table 2, Entry 2). By increasing the amount of *p*-TsOH from 0.5 mmol to 1.0 mmol the required reaction time was shortened from 4 h to 2 min and a remarkable increasing was also observed for product yield (Table 2, Entry 3). Further addition of an amount of *p*-TsOH had no effect on the deprotection of model substrate under solid state conditions (Table 2, Entry 4). Therefore, the equimolar of *p*-TsOH and model substrate was optimum molar ratio.

Table 2. Effect of p-TsOH amounts on deprotection of imine 1a at r.t[a]

Entry	***p*-TsOH (mmol)**	**Time (min)**	**Yield (%)[b]**
1	0.1	240	trace
2	0.5	240	50
3	1.0	2	100
4	1.5	2	100

[a] All reactions were run with imine **1a** (1 mmol) in solid state at r.t.
[b] Isolated yields

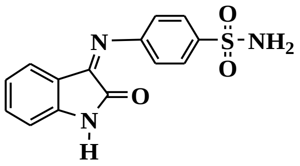
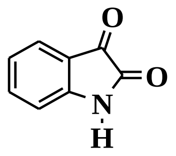
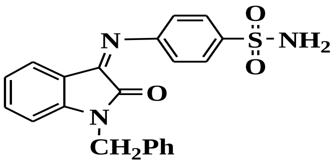
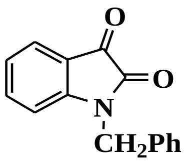
The following mechanism can be written to this reaction (Scheme 2).

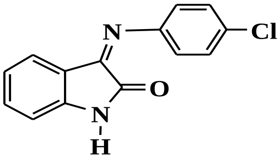
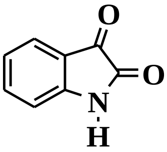
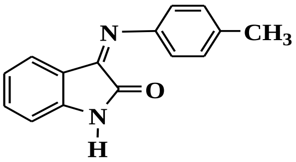
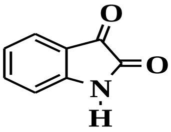
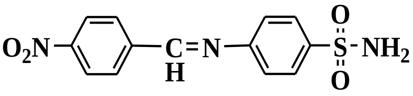
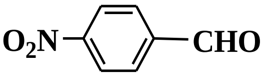
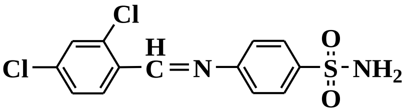
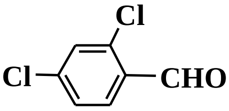
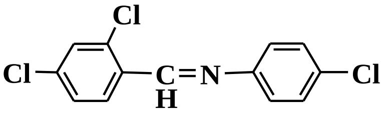
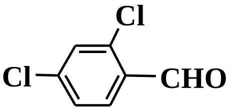
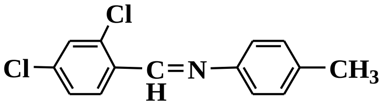
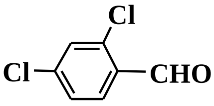
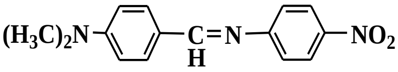
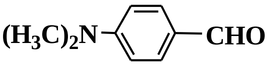
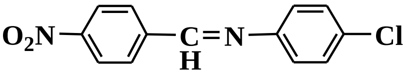
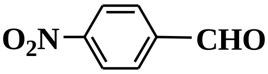
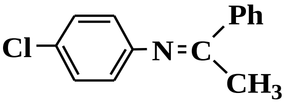
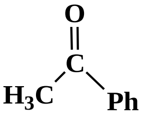
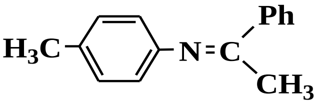
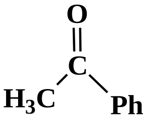
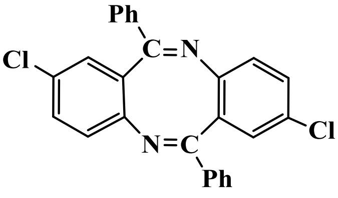
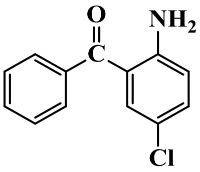


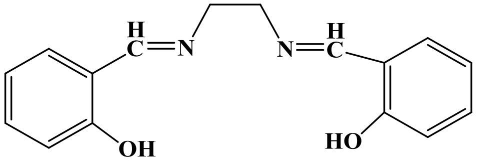
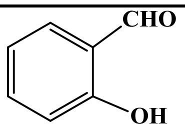
Scheme 2. Plausible mechanism for deimination of imines

The best conditions above (molar ratio 1:1 for substrate/ *p*-TsOH, under solid state condition at r.t) was used for the deprotection of a wide range of imines. The results of this study were summarized in Table 3. Some of the substrates immediately provided the carbonyl compounds in high yield at room temperature (Table 3, Entries 1-6), but some of them required a long time for deprotection at the same temperature that surprisingly elevated the reaction temperature to 45 ºC markedly increased the reaction rate (Table 3, Entries 7-14). For example, when the imine 1k was subjected to the model reaction conditions, acetophenone formed in 30% yield after 10 h at room temperature, while with arising the temperature to 45 ºC this product was obtained in 78% yield within 30 min.

Table 3. The cleavage of C=N bond of imine groups using p-TsOH at r.t in the solid state conditions[a]

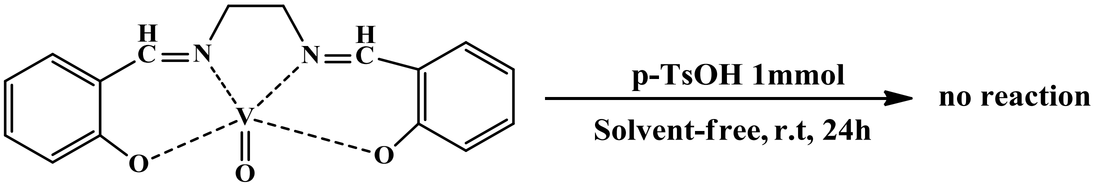
Entry	Substrate	Carbonyl product[b]	Time (min)	Yield(%)[c]
1	 1a	 2a	2	90
2	 1b	 2b	2	8

3	 1c	 2a	6	89
4	 1d	 2a	4	95
5	 1e	 2e	8	80
6	 1f	 2f	10	78
7	 1g	 2f	480 (20)[d]	74 (81)[d]
8	 1h	 2f	480 (20)[d]	٦٥ (٧٦)[d]
9	 1i	 2i	360 (20)[d]	٧٦ (٨٥)[d]
10	 1j	 2e	240 (20)[d]	٨٠ (٨٢)[d]
11	 1k	 2k	600 (30)[d]	٦٥ (٧٨)[d]
12	 1l	 2k	600 (20)[d]	٧٠ (٧٤)[d]
13	 1m	 2m	720 (20)[d]	٦٤ (٨٩)[d]

14	 1n	 2n	360 (20)[d]	۸٤ (۸۸)[d]

[a]Conditions: Imine (1 mmol), *p*-TsOH (1 mmol), in solid state at r.t. [b]All of products are known and were characterized by both comparison of their physical and spectroscopic data (^{1}H NMR, ^{13}C NMR, IR) with those of authentic samples and also by the conversion of them to their corresponding starting materials [21-23]. [c]Isolated yields. [d]The reaction was conducted at 45 ºC.

The stability of imines can be increased if the free electron pair from the nitrogen atom is coordinated to metal. In the other study, the deprotection of imine group in both salen-vanadium oxide complex and salen molecule was investigated. Thus, when a mixture of fine powdered salen (1mmol) and *p*-TsOH (2 mmol) was kept at 45 ºC, the salicylaldehyde was obtained in 88% yield after 20 min (Table 3, Entry 14). In contrast, repeating the reaction with complex of salen-vanadium oxide (1 mmol) and *p*-TsOH (1 mmol) did not show any progress for this reaction even after 24 h and the complex remained unchanged under the same condition (Scheme 3).



Scheme 3. Try to deprotection of imine group in salen-vanadium oxide complex and salen molecule by p-TsOH

Conclusion

In summary, we described a mild, efficient and eco-friendly method for the cleavage of C=N bond of imines using *p*-TsOH in solid state conditions. This reaction was unsuccessful for cleavage of C=N bond of imine groups in complex of salen-vanadium oxide.

Acknowledgments

Authors are grateful of University of Mohaghegh Ardabili for the financial support and the laboratories of Tehran University for the product analysis.

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