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One-pot synthesis of 2-substituted quinoxalines using K10-montmorillonite as heterogeneous catalyst

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ABSTRACT

An efficient one-pot synthesis of 2-substituted quinoxalines from 1,2-diamines and phenacyl bromides is developed using K10-montmorillonite (K10 clay) as a catalyst at 50 °C in acetonitrile medium. This method offers an easy route for the synthesis of substituted quinoxalines in high yields. A plausible mechanism is proposed in which quinoxalines are formed via dehydration–dehydrohalogenation–cyclization sequence. Further, the K10 clay catalyst is recovered by simple filtration and reused six times without any loss in its catalytic activity.

Synthesis of quinoxalines and their derivatives is receiving considerable attention due to their biological activities and pharmaceutical applications.[1] Quinoxalines have been used in anticancer,[2] antiviral,[3] antibiotic (echinomycin, leromycin and actinomycin) anti-inflammatory, and kinase inhibition activities.[4] Furthermore, they are also used as potential building blocks for the synthesis of organic semiconductors,[5] electroluminescent materials,[6] cavitands,[7] dehydroannulenes,[8] and dyes.[9]

A number of methods have been developed for the synthesis of quinoxalines. One such method involves the condensation of a 1,2-diketone with aryl-1,2-diamine to give the corresponding quinoxaline at refluxing temperature in ethanol, benzene, or acetic acid.[10] This reaction is catalyzed by iodine,[11] indium(III) chloride,[12] copper(II) sulfate,[13] ceric ammonium nitrate,[14] *o*-iodoxybenzoic acid,[15] phosphorus oxychloride,[16] silica gel,[17] gallium(III) triflate,[18a] and clayzic.[18b] On the other hand, 1,4-addition of the 1,2-diamines to diazenylbutenes[19] and oxidation of α-hydroxyketones with 1,2-diamines[20] have also been reported for the synthesis of the corresponding quinoxalines. Also, the synthesis of quinoxaline has been reported by the reaction of 1,2-diamines with substituted phenacyl bromides in solid phase,[21] and using catalysts like perchloric acid supported on silica,[22a] trimethylsilyl chloride,[22b] KF-alumina,[22c] β-cyclodextrin,[23] and 1,4-diazabicyclo [2,2,2]octane.[24] However, these reported procedures suffer from limitations such as the need of excess catalyst loading, toxic reagents, and the requirement of high temperature to conduct the reaction. In particular, acetic acid and perchloric acid are more hazardous and the current trend is to develop green and sustainable protocols toward the same.

In this context, considerable attention has been devoted to perform organic transformations using solid acids as heterogeneous catalysts.[25] In the present work, we report a new synthetic strategy for the synthesis of quinoxaline derivatives catalyzed by K10 clay under mild reaction conditions. K10 clay is a layered alumina-silicate clay with a dioctahedral layer sandwiched between two tetrahedral layers.[26] K10 clay with a moderate acid strength and a surface area of 250 m^2/g can be used as heterogeneous catalyst for various organic transformations due to their tunable Bronsted and Lewis acidities[27a,b] and is considered to be an environmentally benign solid acid catalyst that offers several advantages, such as ease of handling, non-corrosiveness, low cost, and reusability.

The reaction conditions were optimized[28] for the synthesis of 2-phenylquinoxaline using *o*-phenylenediamine and phenacyl bromide as model substrates and the observed results are given in Table 1. Blank control experiments in the absence of catalyst showed 8% and 17% of product at room temperature and 50 °C in 8 and 3 h respectively in acetonitrile medium (Table 1, entries 1 and 2). In the absence of solvent, K 10 clay gave 13% and 69% yields at room temperature and 50 °C respectively in 3 h (Table 1, entries 3 and 4). Further increasing the reaction time to 6 h at 50 °C improved the yield to 77% (Table 1, entry 5) and it can be considered as an encouraging result, from the green and sustainable process since it does not require any solvent in the reaction

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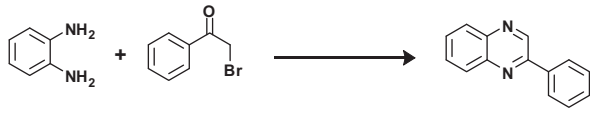
Table 1
Optimization of reaction conditions for the synthesis of 2-phenylquinoxaline[a]



Entry	Catalyst	Solvent	Time (h)	Temp (°C)	Yield[b] (%)
1	—	ACN	8	rt	8
2	—	ACN	3	50	17
3	K10 clay	None	3	rt	13
4	K10 clay	None	3	50	69
5	K10 clay	None	6	50	77
6	K10 clay	ACN	3	rt	33
7	K10 clay	ACN	8	rt	90
8	K10 clay	ACN	0.5	50	18
9	K10 clay	ACN	1	50	40
10	K10 clay	ACN	2	50	71
11	K10 clay	ACN	3	50	92
12	K10 clay	Methanol	0.5	50	10
13	K10 clay	Methanol	1	50	29
14	K10 clay	Methanol	2	50	56
15	K10 clay	Methanol	3	50	80
16	K10 clay	Methanol	6	50	82
17	K10 clay	THF	3	50	85
18	K10 clay	Chloroform	3	50	75
19	K10 clay	Acetone	3	50	70
20	K10 clay	Dichloromethane	3	50	68
21	K10 clay	DMF	3	50	59
22	K10 clay	Benzene	3	50	60
23	K10 clay	Toluene	3	50	62

[a] Reaction conditions: *o*-phenylenediamine (1 mmol), phenacyl bromide (1 mmol), K10 clay (50 mg), solvent (5 mL).
[b] Isolated yield.

process. K10 clay showed highest yield of 92% in acetonitrile in 3 h while methanol gave 80% under identical conditions (Fig. 1) and no further increase in yield was noticed even after 6 h (Table 1, entries 8–16). Further, the reactions were performed using acetonitrile as a solvent only from the perspective of yield but however performing reactions in methanol or under solvent-less conditions is potentially more interesting from green and sustainable processes points of view. On the other hand, 90% yield (Table 1, entry 7) of the desired product was obtained in acetonitrile at room temperature in 8 h. Moderate to high yields were achieved with conventional organic solvents (Table 1, entries 17–23).

On the other hand, efforts were also made to optimize the catalyst loading in this reaction and the observed results are given in Fig. 2. It can be seen very clearly that the percentage yield gradually increased from 15 to 50 and reaches a maximum at 75 mg of

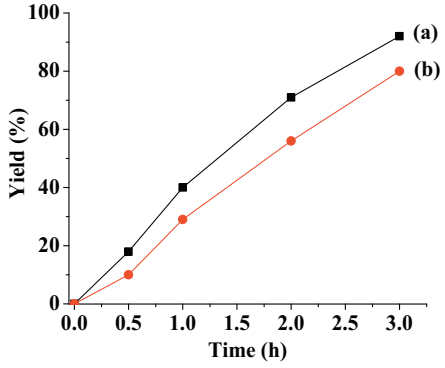


Figure 1. Time conversion plot for the synthesis of 2-phenylquinoxaline in acetonitrile (a) and methanol (b); reaction conditions: *o*-phenylenediamine (1 mmol), phenacyl bromide (1 mmol), K10 clay (50 mg), solvent (5 mL), 50 °C.

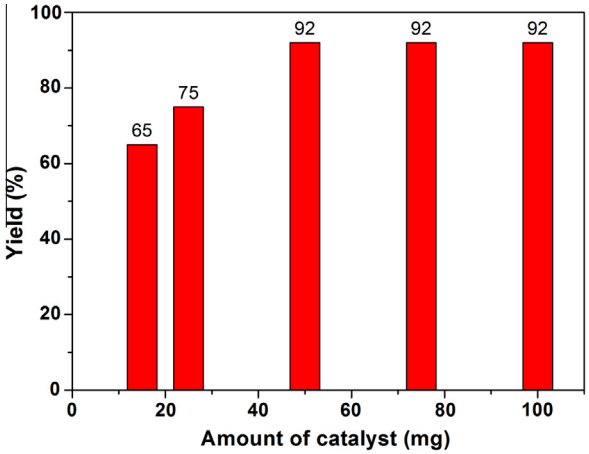


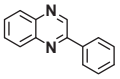
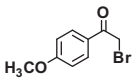
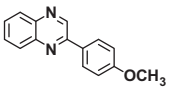
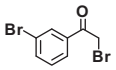
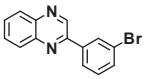
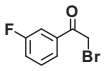
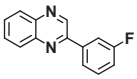
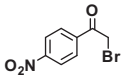
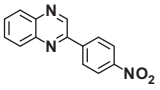
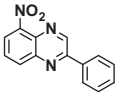
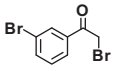
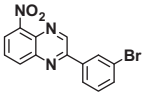
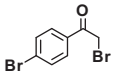
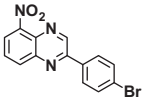
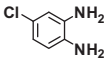
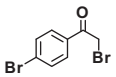
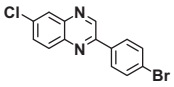
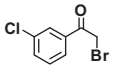
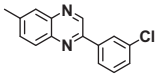
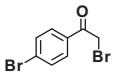
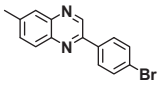
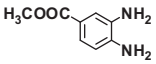
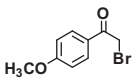
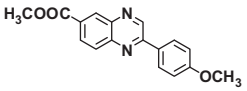
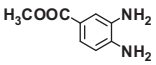
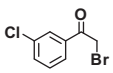
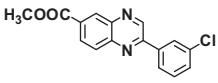
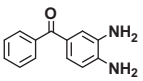
Figure 2. Effect of catalyst loading on the yield of 2-phenyl quinoxaline. Reaction conditions: *o*-phenylenediamine (1 mmol), phenacyl bromide (1 mmol), K10 clay, CH_3CN (5 mL), 50 °C, 3 h.

catalyst loading. Further increase of catalyst amount fails to increase the yield suggesting an optimal value of catalyst loading for this reaction under the present experimental conditions.

These interesting results observed in the synthesis of 2-phenylquinoxaline in high yield under mild reaction conditions encouraged us to check the generality of this methodology using a wide variety of 1,2-diamines and phenacyl bromides. Under the optimized conditions, both electron rich and electron deficient substituents on diamines and phenacyl bromides resulted in high yield of quinoxaline derivatives (Table 2). For example, the reaction of *o*-phenlyenediamine with phenacyl bromide containing electron-donating (4-OMe) and electron-withdrawing (3-Br, 3-F, 4-NO_2) substituents resulted in high yields of substituted quinoxalines (Table 2, entries 2–5). On the other hand, substituted *o*-phenylenediamines also gave the corresponding 2-substituted quinoxalines with various phenacyl bromides in high yields (Table 2, entries 6–13). Increasing the molecular dimension of 1,2-diamine as in the case of 3,4-diaminobenzophenones, (Table 2, entries 14–17) with various phenacyl bromides has also resulted in very high yield of the desired products. It is also interesting to note that condensation between the ketone and the amino group is not observed under the present experimental conditions. It is very clear from the observed results that the nature of the substituents, steric hindrance, and molecular dimension of the reactants have a minimal role to play in determining the rate of reaction and as a result, most of the reactions afforded very high yields under mild reaction conditions. Thus, the operational simplicity of the process, achieving the desired products in high yields regardless of the kind of starting materials, low cost of the catalyst, and the use of acetonitrile as solvent make this protocol an alternative green chemical process for the already existing methodologies.

One of the main advantages of using heterogeneous catalysts such as K10 clay is that they can be recovered and reused efficiently. In this context, the reusability of K10 clay was checked in the synthesis of 2-phenylquinoxaline under optimized conditions, as given in Table 1. After completion of the reaction, the catalyst was recovered by filtration, washed thoroughly with acetonitrile and dried at 80 °C for 3 h. The recovered catalyst was used for consecutive runs under identical reaction conditions and the observed results are given in Table 3. Hence, the catalyst exhibited high activity up to six reuses with only a marginal decrease in the yield which may be caused from the loss of catalyst during recycling or adsorption of organic products over the catalyst by poisoning their active sites. In any case, this catalyst exhibited high potential by maintaining its activity while recycling experiments.

Table 2
Synthesis of various substituted quinoxalines in the presence of K10 clay as heterogeneous catalyst[a]

Entry	Diamine	Phenacyl bromide	Quinoxaline	Yield[b] (%)
1				92
2				90
3				95
4				91
5				93
6				89
7				85
8				86
9				87
10				91
11				93
12				87
13				89
14				83
15				86
16				87
17				85

[a] Reaction conditions: diamine (1 mmol), phenacyl bromide (1 mmol), K10 clay (50 mg), acetonitrile (5 mL), 50 °C, 3 h.
[b] Isolated yield.

The surface acidity of natural clays with Na^+ and NH_4^+ as interstitial cations ranges from +1.5 to −3 on Hammett scale.[27c] The presence of Lewis and Bronsted acid sites has been reported earlier by IR studies using adsorption of ammonia as a probe molecule. Two absorption bands have been observed at 1445 and 1630 cm^{-1}. The band at 1630 cm^{-1} was attributed to the bending

Table 3
Reusability of K10 clay in the synthesis of 2-phenylquinoxaline[a]

Run	1st	2nd	3rd	4th	5th	6th
Yield[b] (%)	90	88	87	85	83	82

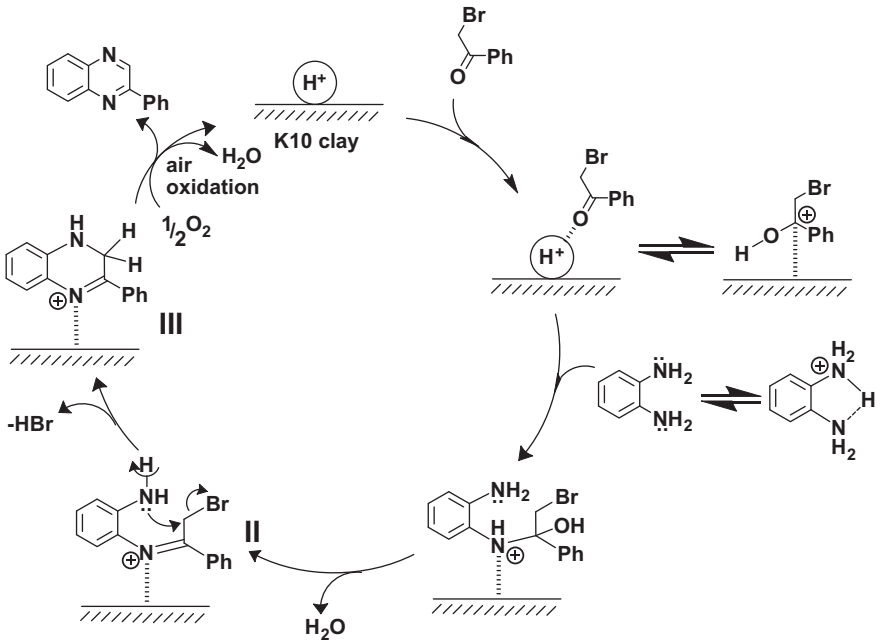
[a] Reaction conditions: *o*-phenyleneamine (1 mmol), phenacyl bromide (1 mmol), K10 clay (50 mg), acetonitrile (5 mL), 50 °C, 3 h.
[b] Isolated yield.

vibration of ammonia coordinatively linked to a Lewis acid centre. The absorption band in the region at 1445 cm^{-1} was assigned to the vibration of the NH_4^+ ion indicating the presence of Bronsted acidic sites.[27d]

Based on the understanding about the different types of acid sites in K 10 clay, a tentative mechanism is proposed involving the reaction of diamine and phenacyl bromide. K10 clay has been used as solid acid catalysts due to the existence of both Lewis and Bronsted acid[29] sites. In the proposed mechanism, K10 clay acting as a Bronsted acid polarizes the carbonyl group of phenacyl bromide giving a carbocationic intermediate as shown in Scheme 1, subsequent nucleophilic attack by *o*-phenylenediamine followed by dehydration gives an imine intermediate **II**. This is followed by the S_N2 type attack by the second amino group leading to dehydrohalogenation and cyclization, giving dihydroquionoxaline **III**. Subsequent clay promoted dehydrogenation gives quinoxaline, with aromatization as the driving force. The clay catalyst is released for further cycle of reaction.

The activity and efficiency of the present work are compared with the reported catalytic systems for the synthesis of 2-substituted quinoxaline (Table 4). Solid phase synthesis of quinoxaline using polymer as a support required multistep synthesis and no data were found on reusability. Although KF-alumina showed higher yields under solvent-less conditions at room temperature, it required high catalyst loading (0.5 g) and its reusability was not reported. In another precedent, β-cyclodextrin was used as catalyst at 70 °C in water but however reusability of catalyst was not reported. The present catalytic system reports high yield of the product at moderate reaction temperature (50 °C) in acetonitrile and it can be reused up to six times which were lacking in the above examples.

In conclusion, we have developed an efficient one-pot synthesis of quinoxaline derivatives from substituted *o*-phenylenediamines and phenacyl bromides in the presence of K10 clay in acetonitrile at 50 °C. This methodology allowed us to synthesize a series of substituted quinoxaline analogues in high yields irrespective of the kind of functional groups in the starting reagents and above all the reaction conditions are milder and greener. The other advantages of this strategy are low cost of catalyst, achieving good yield of products in high purity, operational simplicity, milder



Scheme 1. Proposed mechanism for the formation of 2-phenylquinoxaline catalyzed by K10 clay.

Table 4
Comparison of the present catalytic system with the earlier reported procedures for the synthesis of 2-substituted quinoxaline from substituted *o*-phenylenediamine and phenacyl bromide

Entry	Catalyst (weight)	Solvent	Temp (°C)	Time	Yield (%)	Reusability	Ref.
1	Wang resin as polymer support (100 mg)	—[a]	—[a]	—[a]	78	NR	21b
2	Perchloric acid supported on silica (50 mg)	Acetonitrile	rt	15–60 min	80–95	3[b]	22a
3	Trimethylsilyl chloride (1 mmol)	Water	70	8 h	57–91	NR	22b
4	KF-alumina (0.5 g)	None	rt	1.5–5 h	82–94	NR	22c
5	β-Cyclodextrin (1 mmol)	Water	70	2–2.5 h	83–92	NR	23
6	1,4-Diazabicyclo[2,2,2] octane (20 mol %)	THF	rt	20–50 min	84–93	NR	24
7	K10 clay (50 mg)	Acetonitrile	50	3 h	83–95	6(82)	Present system

[a] Required different solvents, temperatures and time due to multistep.
[b] Data were not reported; NR stands for Not Reported.

reaction conditions, eco-friendliness, and the high potential in the catalyst reuse without significant changes in yield up to six runs. These salient features make this protocol to be environmentally benign and viable for industrial applications.

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Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.tetlet.2014.01.087.

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28. *General procedure for the synthesis of 2-substituted quinoxaline derivatives:* Substituted 1,2-phenylenediamines (1 mmol) and substituted phenacyl bromides (1 mmol) were dissolved in acetonitrile (5 mL). To this solution, K10 clay (50 mg) was added and heated at 50 °C for 3 h. After completion, the reaction mixture was filtered to remove the catalyst and washed with ethyl acetate (10 mL). The filtrate was concentrated and the residue was purified by column chromatography using hexane/ethylacetate (15% ethyl acetate in hexane) as an eluent to afford quinoxaline derivatives in high yields. The recovered catalyst was thoroughly washed with ethyl acetate, air dried, activated at 80 °C for 3 h and reused for successive runs.
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