Synthesis of benzoimidazoquinazolinone and indolylxanthenone derivatives using Keggin-type heteropoly-11-molybdo-1-vanadophosphoric acid supported on Montmorillonite K-10 clay as catalyst: a green approach

Antony Muthu Prasanna¹,² · Murugan Kumaresan³ · Karuppaiah Selvakumar⁴ · Meenakshisundaram Swaminathan⁵ · Ponnusamy Sami¹

Received: 15 August 2022 / Accepted: 12 October 2022 / Published online: 25 October 2022
© The Author(s), under exclusive licence to Springer Nature B.V. 2022

Abstract
Biologically as well as medicinally important two different organic scaffolds, viz. benzimidazole and quinazoline, are present in the class of heterocyclic compounds called benzoimidazoquinazolinone. Similarly, indolylxanthenones are the compounds containing two important organic moieties such as indole and xanthene. In this work, a new green protocol for the synthesis of benzoimidazoquinazolinone and indolylxanthenone derivatives was attained under environmental-friendly solvent-free condition through a simple one-pot three-component condensation reaction. This condensation was achieved by using 10% heteropoly-11-molybdo-1-vanadophosphoric acid (H₄[PVMo₁₁O₄₀])-loaded Montmorillonite K-10 clay material (PVMoK-10) as an efficient heterogeneous catalyst. The identification and characterization of the derivatives were done by physical as well as spectral techniques. Synthesis of ten derivatives of benzo[4,5]imidazo[2,1-b]quinazolin-1(2H)-one and two derivatives of 9-(1H-indol-3-yl)-2,3,4,9-tetrahydro-1H-xanthen-1-one was successfully achieved using this protocol. A tentative reaction mechanism has also been proposed for the synthetic plans.

Keywords One-pot multi-component synthesis · Heteropoly acid-supported Montmorillonite K-10 clay catalyst · Benzoimidazoquinazolinone · Indolylxanthenone
Introduction

Heterocyclic compounds are renowned for their diversified and potential biological activity essential for everyday life [1–6]. More specifically, the nitrogen-containing heterocyclic moieties are more active in the biological and the pharmacological areas [7–9] because of their resemblance of structure with some naturally existing organic molecules, which shows familiar biological activity [4]. Furthermore, the review of literature reveals that polyheterocyclic molecular entity enhances the biological versatility of the natural product [10]. This part of the work dealt with the synthesis of two such types of polyheterocyclic compounds, viz., benzoimidazoquinazolinones and indolylxanthenones.

Benzoimidazoquinazolinone has been attracted extensively by synthetic organic as well as medicinal chemists, due to their diversified applications in the multitude of areas [5, 11–13]. On the other hand, the chromene part of xanthene present in indolylxanthenones is one of the more privileged scaffolds in natural products [14]. They are found to be a valuable precedence motif for designing pharmacophore in the area of medicinal chemistry for the development of drugs [15–17]. Organic chemists are more concerned with devising synthetic strategies for the synthesis of such biologically active organic molecules. In this regard, multi-component reactions (MCRs) become an appropriate method for the synthesis of variety of medicinally and biologically important heterocyclic organic compounds. MCR involves the selective production of desired complex organic compound in high yield at short reaction time by taking two or more number of readily available starting materials called the synthetic organic precursors in a single pot. MCR becomes a powerful tool for the synthesis of complex organic compounds that are used as drugs through convergent synthetic approach. MCR enables the production of a variety of poly-functionalized organic heterocyclic drug-like compounds. Hence, in contemporary times evolution of new MCR procedures using different types of catalysts has received much attention in synthetic organic chemistry. MCRs diligently have been admired due to their powerful upshot on drug innovation for their sharp focus in shortening the synthetic pathway of drugs and their derivative products [18–25]. Polyoxometalates (POMs) a class of oligomeric aggregates of metal cations bridged by oxide anions capable of existing as free solid acid materials, heteropoly acids (HPAs) attract recent attention in the field of solid acid catalysis. HPAs have an ability to act as proton donor and thereby activate many organic transformations. The main disadvantage of HPA is its low surface area and separation problem from reaction mixture due to very high solubility of HPA in the reaction medium. In order to overcome these problems, HPAs supported on several solid supports and variety of organic transformations have been carried out using these catalytic materials. Tayebee et al. [26–36] have successfully attempted many novel protocols to synthesize biologically active organic molecules using heteropolyoxometalate-supported catalysts through MCRs. Mohammad Nikpassand et al. evolved the usage of modified zeolite solid acid catalysis for the synthesis of variety of organic compounds using MCR protocols [37–44]. This research work is one such attempt to develop a new
synthetic protocol for the synthesis of Benzoimidazoquinazolinone and indolylxanthenones using heteropolyoxometalate-supported Montmorillonite K-10 clay catalyst under environmentally benign reaction conditions.

A review of the literature finds that a few synthetic approaches have been reported for the synthesis of benzimidazoquinazolines including its synthesis through a three-component condensation of 2-aminobenzimidazole, 1,3-cyclohexadione and substituted aromatic aldehydes in the presence of catalyst silica gel under solvent-free condition [45]. Ali Maleki et al. [46] reported the synthesis of benzimidazo[2,3-b]quinazolinone derivatives through a one-pot multi-component reaction promoted by a chitosan-based composite magnetic nanocatalyst. Ziarani et al. [47] proposed a synthetic route for the synthesis of benzimidazoquinazolinones using nanoporous Santa Barbara Amorphous-15 silica (SBA-15).

We functionalized sulfonic acid (SBA-Pr-SO$_3$H) as heterogeneous acid catalyst via the reaction of substituted aldehydes with a cyclic 1,3-diketone and 2-aminobenzimidazole under solvent-free condition. Heravi et al. [48] proposed the synthesis of triazoloquinazolinones and benzimidazoquinazolinones by the condensation reaction of amine sources like 3-amino-1,2,4-triazole/2-amino benzimidazole with aldehydes and dimedone using acetonitrile-mediated sulfamic acid catalyst under reflux condition. Mousavi et al. [49] proposed synthesis of derivatives of [1, 2, 4] triazolo[5,1-b] quinazolin-8(4H)-one and hexahydro[4,5] benzimidazo[2,1-b]quinazolinone by the reaction of substituted aldehydes with dimedone and 3-amino-1,2,4-triazole/2-aminobenzimidazole using acetic acid promoter under metal-free condition at 60 °C. Mousavi et al. [50] developed an eco-friendly synthesis of triazoloquinazolinones and benzimidazoquinazolinones derivatives via the condensation reaction of substituted aromatic aldehydes with cyclic 1,3-diketone and 3-amino-1,2,4-triazole/2-amino benzimidazole by heterogeneous nano-SiO$_2$ catalyst in acetonitrile medium at room temperature. Yao et al. [51] accomplished the synthesis of 4-aryl-1H-pyrimido[1,2-a] benzimidazole derivatives by the condensation of substituted aryl aldehyde and 2-aminobenzimidazole with 1,3-dicarboxyl compounds in 1-butyl-3-methyl imidazolium tetrafluoroborate ([bmim$^+$][BF$_4$]$^-$) ionic liquid medium at 90 °C. Mousavi et al. [52] proposed the synthesis of triazoloquinazolinone and benzimidazoquinazolinone derivatives via the condensation reaction of different aryl aldehydes and cyclic 1,3-dicarboxyl compound with 2-aminobenzimidazole/3-amino-1,2,4-triazole catalyzed by p-toluenesulfonic acid monohydrate at 40–50 °C in acetonitrile reaction medium. Puligoundla et al. [53] reported the synthesis of benzimidazoloquinazolinone derivatives through the condensation of an amino source, 2-aminobenzimidazole, aryl aldehyde and dimedone. The reaction was mediated by molecular iodine in acetonitrile solvent medium under reflux condition for about 15 min. Heravi et al.[54] developed the synthesis of [1, 2, 4]triazolo/benzimidazoquinazolinone derivatives by refluxing 3-amino-1,2,4-triazole/2-amino benzimidazole with dimedone and aromatic aldehyde using Wells–Dawson-type heteropoly acid, $H_6[P_2W_{18}O_{62}]18H_2O$ as catalyst in acetonitrile solvent medium. Petrova et al. [55] developed a three-component condensation reaction of arylglyoxals, 2-aminobenzimidazole and 1,3-cyclohexanedione under conventional as well as microwave conditions. Mourad et al.
[56] reported the microwave-mediated synthesis of benzimidazoquinazolinones via the reaction of 2-aminobenzimidazole with aryl aldehydes and dinedone in dimethylformamide. Kasaralikar et al. [57] reported the synthesis of oxochromenyl xanthenone/indolyl xanthenone derivatives via the condensation of various substituted salicylaldehydes and 4-hydroxy coumarin/indole with dinedone at room temperature using an ionic liquid 1-hexyl-3-methylimidazolium hydrogen sulfate, ([Hmim]HSO₄) catalyst in ethanolic medium at 50 °C. Bhattacharjee et al. [58] performed the synthesis of 9-(1H-Indol-3-yl)-3,3-dimethyl-2,3,4,9-tetrahydro-1H-xanthen-1-one using ammonium chloride in aqueous medium via a condensation reaction of salicylaldehyde with cyclic 1,3-diketones and indole. Ganguly et al. [59] accomplished L-proline-mediated coupling reaction of 2-hydroxybenzaldehyde with indole and 5,5-dimethyl-1,3-cyclohexanedi one for the synthesis of 9-(1H-indol-3-yl)-xanthen-4-(9H)-ones under micellar condition using sodium dodecyl sulfate in water at ambient temperature.

In spite of their pre-eminence of the aforementioned synthetic protocols, the synthesis of benzimidazoquinazolines and indolylxanthenone is perceived to be deprived of easy, simple, greener and direct methods. The discovery of simpler and greener method for the synthesis is of utmost significance and is essential for overcoming these difficulties by the emergence of a simpler and greener method. Subsequently, it is planned to prepare benzimidazoquinazolines and indolylxanthenone in new methodology. In continuation of our successful previous green synthetic protocol for the synthesis of several organic heterocyclic compounds [60–64], herein, we devised a simple and economical strategy to synthesize benzo[4,5]imidazo[2,1-b]quinazolin-1(2H)-one derivatives (R1) by condensation of 1,3-cyclohexadione with 2-aminobenzimidazole, and various substituted aldehydes and 9-(1H-indol-3-yl)-2,3,4,9-tetrahydro-1H-xanthen-1-one derivatives (R2) through the condensation reaction of indole, 1,3-cyclohexadione and salicylaldehyde at 100 °C under solvent-free medium using a heterogeneous catalyst heteropoly-11-molybdo-1-vanadophosphoric acid, H₄[PVMo₁₁O₄₀] supported on the clay material Montmorillonite K-10 clay (PVMoK-10).

**Experimental**

**Materials and methods**

1,3-Cyclohexadione, 2-aminobenzimidazole, indole, different substituted aldehydes and Montmorillonite K-10 clay (Mont-K10) were purchased from Sigma-Aldrich, and all were used as such without further purification. The 12-heteropoly acid (HPA) H₃[PMo₁₂O₄₀], its mono-vanadium (V)-substituted HPA, H₄[PVMo₁₁O₄₀], di-vanadium (V)-substituted HPA, H₅[PV₂Mo₁₀O₄₀] and the HPA-supported Mont-K10 with different loadings of HPA, viz. 10%, 20% and 30% in (w/w), were prepared and characterized using the procedures reported in the literature [65–67].
General reaction procedure for the synthesis of benzo[4,5]imidazo[2,1-b]quinazolin-1(2H)-one derivatives (R1)

Benzo [4, 5] imidazo[2,1-b]quinazolin-1(2H)-one derivatives were prepared as per the procedure adopted in the literature [48, 68]. A mixture of one millimole each of 2-aminobenzimidazole, 1,3-cyclohexadione, substituted aromatic aldehyde and 0.05 g of the catalyst 10% PVMoK-10 were heated at 100 °C for 1 h in oil bath. TLC method using petroleum ether—ethyl acetate solvent mixture in the ratio of 7:3 was used to follow the proceeding of the conversion. After the completion of the reaction was ascertained through TLC, 5 mL of ethanol was added to the reaction mixture and the mixture was heated. The organic product thus formed was soluble in ethanol. The solid catalyst from the reaction mixture was removed by filtration. The filtrate was then poured into crushed ice pieces, and the solid product isolated was filtered out. The product formed was washed thoroughly with water and dried and recrystallized from ethanol. The recovered catalyst by filtration method was thoroughly washed with water and dried in air oven for about an hour at 100 °C.

General reaction procedure for the synthesis of 9-(1H-indol-3-yl)-2,3,4,9-tetrahydro-1H-xanthen-1-one derivatives (R2)

9-(1H-indol-3-yl)-2,3,4,9-tetrahydro-1H-xanthen-1-one derivatives were prepared as per the procedure reported in the literature [68–70]. One millimole each of indole, 1,3-cyclohexadione, salicylaldehyde were mixed with 0.05 g of 10% PVMoK-10 and heated for about 1 h at 100 °C in an oil bath. The completion of the reaction was followed by TLC using the eluent ethyl acetate and n-hexane solvent in the ratio of 7:3. After the reaction is over, 5 mL of ethanol was added in order to dissolve the organic product formed. The solid catalyst from the reaction mixture was filtered out while heating. A solid product was regenerated by pouring the filtrate into water, and the solid separated out was filtered off, washed thoroughly with water, dried well and recrystallized using solvent ethanol. The recovered catalyst by filtration was washed thoroughly with water and dried in air oven at 100 °C for about an hour.

IR Affinity-1 Shimadzu FT-IR spectrophotometer is used for recording Fourier transform infrared (FT-IR) spectra, under atmospheric conditions using KBr pellets. Bruker 400 MHz NMR instrument is utilized for recording proton and 13C NMR with DMSO-d6 as solvent and tetramethylsilane as internal standard. Melting points of samples were noted by the electro-thermal melting point apparatus. Elemental analyses were studied using ElementarVario EL III equipment. High-resolution mass spectra (HRMS) were recorded using the maXis Impact 282,001.00081 Mass Spectrometer for the selected products.
Results and discussion

Optimization of catalyst

The efficacy of the catalyst was investigated by choosing the condensation reaction of 2-aminobenzimidazole, 1,3-cyclohexadione with benzaldehyde in the presence of catalyst as a model reaction for the preparation of R1 and the condensation of indole, 1,3-cyclohexadione and salicylaldehyde in the presence of catalyst for the preparation of the R2. The reactions were strived with various catalytic materials such as raw Mont K-10, HPAs, viz. H$_3$[PMo$_{12}$O$_{40}$] (PMo), H$_4$[PVMo$_{11}$O$_{40}$] (PVMo) and H$_5$[PV$_2$Mo$_{10}$O$_{40}$] (PV$_2$Mo) and 10% HPA-loaded Mont K-10 clay, PVMoK-10 and PV$_2$MoK-10. Product yield in percentages was used to assess the performance of the selected catalyst, and the results are summarized in Table 1. Clearly, the results reveal that the performance of 10% catalysts—PVMoK-10 & PV$_2$Mo-K10—is found to be very high compared to both raw Mont-K10 clay and vanadium-substituted heteropoly acids. Both vanadium (V)-substituted HPA-loaded Mont-K10 clay shows more or less similar catalytic efficacy. Hence, PVMoK-10 has been selected as a catalyst for the current investigation.

Dependency of different loadings of PVMo on Mont K-10 clay

The catalytic performances of PVMo-loaded Mont K-10 for about 5%, 10%, 30% and 40% were evolved for both the reactions as per the reported procedure [50]. The efficiency of the catalysts was evaluated in terms of percentage yield of products (R1 and R2). The results are summarized in Table 2. It is found that the 10% PVMoK-10 showed equivalent efficiency as that of 20% and 30% loading of PVMo on Mont-K10, and hence, 10% PVMoK-10 has been selected as the catalyst to perform the present synthetic transformation.

Table 1  Optimization of catalyst for one-pot three-component synthesis of 12-phenyl-3,4,5,12-tetrahydrobenzo[4,5]imidazo[2,1-b]quinazolin-1(2H)-one and 9-(1H-indol-3-yl)-2,3,4,9-tetrahydro-1H-xanthen-1-one derivative

| Run | Catalyst | Catalyst amount (g) | Yield$^a$ (%) |
|-----|----------|---------------------|---------------|
|     |          |                     | R1  | R2  |
| 1   | Mont-K10 | 0.05                | 43  | 42  | 39  |
| 2   | PMo      | 0.05                | 51  | 48  | 47  |
| 3   | PVMo     | 0.05                | 62  | 54  | 56  |
| 4   | PV$_2$Mo | 0.05                | 65  | 66  | 59  |
| 5   | PMo-K10  | 0.05                | 75  | 74  | 62  |
| 6   | PV$_2$Mo-K10 | 0.05 | 94  | 94  | 88  |
| 7   | PV$_2$Mo-K10 | 0.05 | 95  | 92  | 88  |

Reaction conditions: R1: 2-aminobenzimidazole (1 mmol), 1,3-cyclohexadione (1 mmol) and benzaldehyde (1 mmol), Solvent-free condition, 100 °C, 1 h
R2: Indole (1 mmol), 1,3-cyclohexadione (1 mmol) and salicylaldehyde (1 mmol), Solvent-free condition, 100 °C, 1 h

$^a$Isolated yields
Synthesis of benzoimidazoquinazolinone and indolylxanthenone...

Dependency of solvents

The reactions were run in different solvent media such as EtOH, MeOH, H2O, MeCN, DCE, DMF, CHCl3, 1,4-dioxane, n-hexane and toluene at elevated temperature (refluxing for an hour). The reaction was also attempted under solvent-free reaction condition. The results of the experiments in terms of yield of products R1 and R2 are given in Table 3. The results favor the solvent-free reaction.

### Table 2

| Run | % of loading of PVMo on Mont-K10 | Catalyst amount (g) | Yielda (%) | R1 | R2 |
|-----|----------------------------------|---------------------|-------------|----|----|
| 1   | 5                                | 0.05                | 73          | 71 |    |
| 2   | 10                               | 0.05                | 94          | 88 |    |
| 3   | 20                               | 0.05                | 95          | 88 |    |
| 4   | 30                               | 0.05                | 95          | 89 |    |

Reaction conditions: R1: 2-aminobenzimidazole (1 mmol), 1,3-cyclohexadione (1 mmol) and benzaldehyde (1 mmol), Solvent-free condition, 100 °C, 1 h

R2: Indole (1 mmol), 1,3-cyclohexadione (1 mmol) and salicylaldehyde (1 mmol), Solvent-free condition, 100 °C, 1 h

*Isolated yields

### Table 3

| S. No. | Solvent used | Yield (%) | R1 | R2 |
|--------|--------------|-----------|----|----|
| 1      | EtOH         | 57        | 51 |    |
| 2      | MeOH         | 71        | 64 |    |
| 3      | H2O          | 31        | 27 |    |
| 4      | CH3CN        | 28        | 22 |    |
| 5      | DCE          | 57        | 53 |    |
| 6      | DMF          | 30        | 26 |    |
| 7      | CHCl3        | 39        | 33 |    |
| 8      | 1,4-dioxane  | 32        | 28 |    |
| 9      | n-Hexane     | 40        | 42 |    |
| 10     | Toluene      | 55        | 49 |    |
| 11b    | Solvent-free | 94        | 88 |    |

Reaction conditions: R1: 1,3-cyclohexadione (1 mmol), 2-aminobenzimidazole (1 mmol) and benzaldehyde (1 mmol), Solvent-free condition, 10% PVMo-K10 0.05 g, Stirring at 1 h

R2: 1,3-cyclohexadione(1 mmol), Indole (1 mmol) and salicylaldehyde (1 mmol), Solvent-free condition, 10% PVMo-K10 0.05 g, Stirring at 1 h

*Isolated yields

b100 °C
condition for the present synthetic transformation since the yield of products under this condition is excellent.

**Synthesis of benzimidazoquinazolinone derivatives**

The optimized reaction procedure has been adapted for the preparation of ten derivatives of R1 (4a-4j) using 2-aminobenzimidazole, 1,3-cyclohexadione and ten structurally diverse aldehydes. The reaction pattern is schematically explained in Scheme 1, and the results are consolidated in Table 4.

**Synthesis of indolylxanthenone derivatives**

Similarly, the optimized reaction procedure has been adapted for the preparation of two derivatives of R2 (4k and 4l) using 1,3-cyclohexadione, salicylaldehyde and two different indoles. The reaction pattern is schematically explained in Scheme 2, and the results are consolidated in Table 5.

With the help of above procedure, ten different R1 derivatives (4a-4j) and two different R2 derivatives (4k and 4l) have been synthesized through one-pot reaction condition using 10% PVMoK-10 catalyst in solvent-free reaction condition. The details of specific reactants used, products formed (4a–l) and their percentages of yields are collected in Tables 4 and 5.

Further HRMS (Fig. 1) of optimized reaction product 12-phenyl-3,4,5,12-tetrahydrono[4,5]imidazo[2,1-\(b\)]quinazolin-1(2H)-one (4a) with the molecular ion peak at 315.0428 confirms the formation of the expected product. Analytical as well as the spectral data of the compounds 4a-l are appended as supplementary materials.
Table 4  Condensation between 2-aminobenzimidazole, 1,3-cyclohexadione and aldehydes using 10% PVMoK-10 as catalyst

| Entry | Aldehydes | Product | Yield\(^b\) (%) |
|-------|-----------|---------|----------------|
| 4a    | CHO       | ![Image](image1) | 94          |
| 4b    | CHO       | ![Image](image2) | 92          |
| 4c    | CHO       | ![Image](image3) | 88          |
| 4d    | CHO       | ![Image](image4) | 85          |
| 4e    | CHO       | ![Image](image5) | 91          |
| 4f    | CHO       | ![Image](image6) | 93          |
Table 4 (continued)

| 4g | 4h | 4i | 4j |
|----|----|----|----|
| ![Image of 4g] | ![Image of 4h] | ![Image of 4i] | ![Image of 4j] |

| 89 | 91.2 | 92 | 89 |

| a Reaction conditions: R1 (4a–j): 2-aminobenzimidazole (1 mmol), 1,3-cyclohexadione (1 mmol) and aldehydes (1 mmol), Solvent-free condition, 100 °C, 1 h | b Isolated yields |

Scheme 2  Synthesis of R2 derivatives (4 k and 4 l) using 10% PVMoK-10 as catalyst
Table 5 Condensation between substituted indoles, 1,3-cyclohexadione and salicylaldehyde using 10% PVMoK-10 as catalysta

| Entry | Indoles | Product | Yieldb (%) |
|-------|---------|---------|------------|
| 4k    | ![Indole 4k](image) | ![Product 4k](image) | 88 |
| 4l    | ![Indole 4l](image) | ![Product 4l](image) | 91 |

*a Reaction conditions: R2 (4k–l): Indole (1 mmol), 1,3-cyclohexadione (1 mmol) and salicylaldehyde (1 mmol), Solvent-free condition, 100 °C, 1 h
*b Isolated yields

![HRMS of 12-phenyl-3,4,5,12-tetrahydrobenzo[4,5]imidazo[2,1-b]quinazolin-1(2H)-one (4a)](image)

**Tentative mechanism**

The tentative mechanism for the synthesis of R1 derivatives using 10% PVMoK-10 catalyst as a promoter has been arrived based on the mechanism reported by Jolodar and Shirini [71] and is shown in Scheme 3. The catalysis involves the following sequence:

(i) The nucleophilic attack of enol form of 1,3-cyclohexadione on the carbonyl carbon of aldehyde leads to the formation of intermediate (I)
(ii) Dehydration of the intermediate (I) forms intermediate (II).

(iii) The hastening of the nucleophilic attack of the amine group on intermediate (II) leads to the formation of (IV) through the intermediate (III) and

(iv) Dehydration of (IV) leads to the formation of desired product. 10 % loaded solid heteropoly acid, PVMo on Mont K-10 catalytic material, accelerates the organic transformation.

The plausible mechanism for the synthesis of R2 derivatives using 10% PVMoK-10 as a promoter has been arrived as per the mechanisms reported in the literature [57, 58] and is shown in Scheme 4. The reaction sequence is given as follows:

(i) The nucleophilic attack of enol form of 1,3-cyclohexadione on the carbonyl carbon of salicylaldehyde leads to the formation of intermediate (I).

(ii) The nucleophile indole reacts at the benzyldiene double bond of the intermediate (I), which further experiences ring-closure reaction intra-molecularly followed by dehydration and give the desired product.

The primary role of heteropoly acid is a source of proton, which activates the carbonyl group.
Recycling of the catalyst

The separation of the catalyst and its reusability are the two most important criteria for the green catalysts.

In the present investigation, the catalyst from the reaction mixture was separated by simply adding excess ethanol with the fused mass produced at the end of the condensation reaction. The product obtained was dissolved in ethanol, whereas the catalyst was separated from the reaction mixture as insoluble residue, filtered off, washed thoroughly with ethanol and dried in hot air oven for an hour at 120 °C. The catalyst thus recovered was reused under the same reaction conditions. It was observed that the reuse of the catalytic material 10% PVMo-K10 showed gradual decrease in its activity. The consecutive performance of the catalyst thus recovered was examined in terms of its % yield of the products and was found to be sustainable for about five times.

Conclusions

An environmentally benign approach has been devised for one-pot three-component synthesis of ten derivatives of benzimidazoquinazolinone and two derivatives of indolylxanthenone with excellent yield of product (85–94%). The synthesis has been achieved under solvent-free reaction medium, using heteropoly-11-molybdo-1-vanadophosphoric acid-supported Montmorillonite K-10 clay material (10%PVMoK-10) as heterogeneous catalyst. Short reaction duration, excellent yield, reusability of the catalytic material, a simple reaction procedure and solvent-free reaction conditions are the advantages of this protocol. A tentative mechanism has been proposed, and
experimental evidences for the support of the mechanism are the future scope of the work.

**Supplementary Information** The online version contains supplementary material available at https://doi.org/10.1007/s11164-022-04859-7.

**Acknowledgements** The authors thank the Managing Board authorities of Virudhunagar Hindu Nadars’ Senthikumara Nadar College (Autonomous), Virudhunagar-626001, Tamil Nadu, India, for providing infrastructural and research facilities.

**Author contributions** AMP and MK done conceptualization, methodology, investigation, formal analysis, writing—original draft. KS and MS performed data curation and spectral verification. PS was involved in resources, formal analysis and supervision.

**Funding** Not applicable.

**Availability of data and materials** Not applicable.

**Declarations**

**Competing interests** The authors declare no competing interests.

**Ethical approval** This declaration is not applicable.

**Conflict of interest** The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

**References**

1. C. Lamazzi, S. Leonce, B. Pfeiffer, P. Renard, G. Guillaumet, C.W. Rees, T. Besson, Bioorg. Med. Chem. Lett. **10**, 2183 (2000)
2. A.B. Insuasty, H. Torres, J. Quiroga, R. Abonia, R. Rodriguez, M. Nogueras, A. Sanchez, C. Saitz, S.L. Alvarez, S.A. Zacchino, J. Chil. Chem. Soc. **51**, 927 (2006)
3. G.D. Galarcei, R.E. Foncea, A.M. Edwards, H. Pessoamahana, C.D.P. Mahana, R.A. Ebenspergeri, Biol. Res. **41**, 43 (2008)
4. R.W. DeSimone, K.S. Currie, S.A. Mitchell, J.W. Darrow, D.A. Pippin, Comb. Chem. High Throughput Screen. **7**, 473 (2004)
5. Y. Zhang, Z. Chen, Y. Lou, Y. Yu, Eur. J. Med. Chem. **44**, 448 (2009)
6. T. Ishida, T. Suzuki, S. Hirashima, K. Mizutani, A. Yoshida, I. Ando, S. Ikeda, T. Adachi, H. Hashimoto, Bioorg. Med. Chem. Lett. **16**, 1859 (2006)
7. S.K. Sahu, M. Azam, M. Banerjee, S. Acharrya, C.C. Behera, S. Si, J. Braz. Chem. Soc. **19**, 963 (2008)
8. A. Saeed, N. Abbas, U. Florke, J. Braz. Chem. Soc. **18**, 559 (2007)
9. G. Cerchiaro, A.M.C. Ferreira, J. Braz. Chem. Soc. **17**, 1473 (2006)
10. L.N. Vostrova, T.A. Voronina, T.L. Karaseva, S.A. Gernega, E.I. Ivanov, A.M. Kirichenko, M. Totrova, Yu. Pharm. Chem. J. **20**, 404 (1986)
11. T. Herget, M. Freitag, M. Morbitzer, R. Kupfer, T. Stammering, M. Marschall, Antimicrob. Agents Chemotherapy. **48**, 4154 (2004)
12. M. Hranjec, G. Karminski-Zamola, Chem. Ind. **57**, 299 (2008)
13. K.P. Chan, H. Yang, A.S. Hay, J. Polym. Sci. **34**, 1923 (1996)
14. T.C. Lima, A.D.C. Santos, D.T.M. Costa, R.J. Souza, A. Barison, M. Steindel, M.W. Biavatti, R. de Brasileira, Farmacognosia. **25**, 7 (2015)
15. C.V. Subbareddy, S. Sumathi, New J. Chem. **41**, 9388 (2017)
16. N. Vukovic, S. Sukdolak, S. Solujic, N. Niciferovic, Food Chem. **120**, 1011 (2010)
Synthesis of benzoimidazoquinazolinone and indolylxanthenone...
Publisher's Note  Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.

Authors and Affiliations

Antony Muthu Prasanna1,2 · Murugan Kumaresan3 · Karuppaiah Selvakumar4 · Meenakshisundaram Swaminathan5 · Ponnusamy Sami1

1 Department of Chemistry, V.H.N. Senthikumara Nadar College (An autonomous institution affiliated to Madurai Kamaraj University, Madurai), Virudhunagar 626 001, India
2 Department of Chemistry, V.V.Vanniaperumal College for Women, Virudhunagar 626 001, India
3 Department of Chemistry, Nadar Mahajana Sangam S. Vellaichamy Nadar College, Nagamalai, Madurai 625019, India
4 College of Engineering, School of Chemical Engineering, Yeungnam University, 280 Daehak-Ro, Gyeongsan, Gyeongbuk 38541, Republic of Korea
5 Department of Chemistry, Nanomaterials Laboratory, International Research Centre, Kalasalingam Academy of Research and Education (Deemed to Be University), Krishnankoil 626126, India