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La_2O_3 as a very effective double-functional catalyst for the production of 2-amino-4-aryl-4*H*-pyrans and Pyrazolopyranopyrimidines

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ABSTRACT

Lanthanum (III) oxide (La_2O_3) was applied as an efficient bifunctional catalyst to develop a new methodology in the production of 2-amino-4-aryl-4*H*-pyrans and pyrazolopyranopyrimidines in the absence of solvent. In both synthetic protocol, due to the dual functionality of lanthanum (III) oxide (i.e. having basic and acidic Lewis sites), it was very useful and general, and plausible mechanisms were presented in support of this generality and high capability. The current protocol featured environmentally friendly conditions, the ability to recycle and reuse the heterogeneous catalyst up to 6 times, providing products in short times with high yields, easy work-up and no need for difficult steps of catalyst synthesis by using a commercially available catalyst, which makes it an appealing route for the preparation of these derivatives.

***Keywords**: Lanthanum(III) oxide; Dual-functional catalyst; 2-amino-4-aryl-4H-pyrans; Pyrazolopyranopyrimidines; Solvent-free conditions; One-pot; multi-component reaction*

1. Introduction

4*H*-Pyran moiety is of significance as it is a vital scaffold of many drugs and biological active materials, such as antimicrobial [1], antituberculosis[2], antibacterial [3], antifungal [4], antioxidant [5], *pseudomonas aeruginosa* biofilm inhibitor [6], anti-inflammatory [7], anti-diabetic [8], anticancer [9], and anticoagulant [10] agents. They have been also utilized in organic light-emitting diode (OLED) emitters [11], logic gates [12], optical chemosensors [13], dye-sensitized solar cells [14], and fluorescent probes [15]. In the literature, many methodologies have been reported using various catalysts for the synthesis of 2-amino-4-aryl-4*H*-pyrans and pyrazolopyranopyrimidines [8, 16-34]. However, despite the advantages of each of these methods, their use is limited due to problems such as ultrasound [18, 24, 26], the need to apply harsh conditions such as high temperature [23], low efficiency of some derivatives [21, 31], and long reaction time and the use of complex catalysts with difficult production steps [11]. Therefore, there is still a need for new methodologies with high effectiveness.

Multi-component reactions (MCR) and solvent-free conditions are well-known, practical, and useful protocols that have been used for the production of a wide range of organic compounds; their advantages have been mentioned in the literature [35-41].

Metal oxides (alone or mixed with other compounds) have been used as effective catalysts in laboratory and industry owing to good activation of adsorbed compounds, enhancing reaction rate and selectivity, stability under reaction conditions, non-hygroscopic, non-toxicity, dual-functionality (acid and base), easier workup and eco-friendly reaction conditions [42]. Recently, these mineral-compounds catalysts have been

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applied in several organic transformations; e.g. Biginelli and Hantzsch reactions [43], selective deoxygenation of palm oil [44], and esterification reaction [45], and production of pyrazolo-phthalazine-diones [46], pyrazolo-triazole-diones [46], nitro-containing compounds [47], spiro-acridine/indoline derivatives [48], cumyl peroxide [49], 2*H*-indazolo-phthalazine-triones [50], carboacyclic nucleosides [51], *N*-alkyl sulfonamides [52], 1*H*-perimidines [53], amides [54], 3,4-disubstituted isoxazol-5(4*H*)-ones [55], and 9-aryl-1,8-dioxo-octahydroxanthenes [56].

With the aim of minimizing the above problems, here we report how to use lanthanum (III) oxide (La_2O_3) as an efficient, recyclable and accessible double-functional catalyst for the production of 2-amino-4-aryl-4*H*-pyrans and pyrazolopyranopyrimidines through (i) the three-component condensation among aromatic aldehydes (1 mmol), dimedone (1 mmol), and malononitrile (1 mmo;), and (ii) the pseudo-four-component condensation of aromatic aldehydes (1 mmol) with ethyl acetoacetate (1 mmol), hydrazine hydrate (1 mmol) and barbituric acid (1 mmol), respectively, in the absence of solvent and eco-friendly environment.

2. Experimental

2.1. Approach to construct 2-amino-4-aryl-4H-pyrans

In a reaction vessel, a mixture comprising malononitrile (1 mmol, 0.066 g), aldehyde (1 mmol), dimedone (1 mmol, 0.140 g), and La_2O_3 (0.15 mmol, 0.049 g) was stirred at 90°C with a glass rod until TLC confirmed the consuming the reactants (i.e. end of reaction). After this time, the reaction mixture was cooled to room temperature, and diluted with 6 ml of ethyl acetate, and stirred for one min under reflux conditions, then La_2O_3 was separated by filtration. The separated catalyst was dried after washing with ethanol (3×3 mL) under vacuum conditions and prepared for the next run. Finally, pure pyran was acquired by recrystallization of the residue in ethanol (95%) after ethyl acetate distillation.

2.2. Approach to fabricate pyrazolopyranopyrimidines

In a reaction vessel, a mixture comprising barbituric acid (1 mmol, 0.128 g), hydrazine hydrate (1 mmol, 0.032 g), ethyl acetoacetate (1 mmol, 0.130 g), and La_2O_3 (0.15 mmol, 0.049 g) was stirred at 60°C with a glass rod until TLC confirmed the consuming the reactants. After this time, the reaction mixture was cooled to room temperature, and diluted with 8 ml of ethyl acetate, and stirred for one min under reflux conditions, then La_2O_3 was separated by filtration. The separated catalyst was dried after washing with ethanol (3×3 mL) under vacuum conditions and prepared for the next run. Finally, pure pyrazolopyranopyrimidine was acquired by recrystallization of the residue in ethanol (95%) after ethyl acetate distillation.

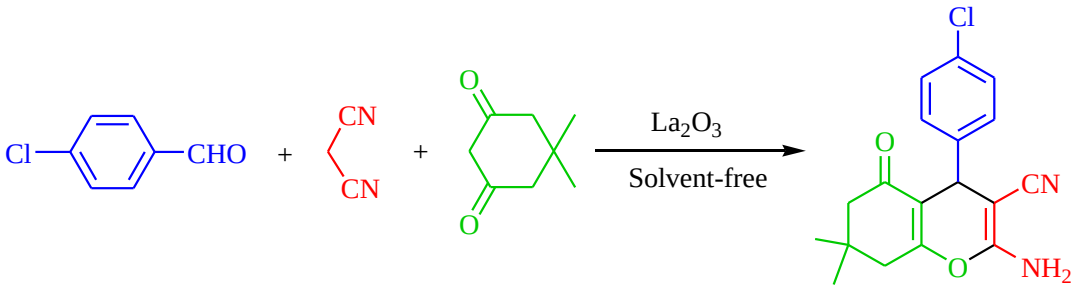
3. Results and Discussion

3.1. Investigating catalytic activity of La_2O_3 in the synthesis of 2-amino-4-aryl-4H-pyrans

At the start, with the aim of preparing 2-amino-4-aryl-4*H*-pyrans under solvent-free conditions, the reaction of dimedone (1 mmol), 4-chlorobenzaldehyde (1 mmol), and malononitrile (1 mmol) was selected as a model (**Scheme 1**), and it was monitored with various amounts of La_2O_3 at a range of 85-95 °C. The results are demonstrated in **Table 1**. According to the table, the optimal state of the reaction is obtained using 0.15 mmol of La_2O_3 at 90 ºC (Table 1, entry 2). It is noteworthy that the reaction failed under catalyst-free conditions even with prolonged heating at 120 °C (**Table 1**, entry 6).

In continue, after determining the optimal conditions, in order to produce different derivatives of 2-amino-4-aryl-4*H*-pyran, different aryl aldehydes (i.e. benzaldehyde and aryl aldehyde substituted with atoms or electron donor or acceptor groups) with malononitrile and dimedone were reacted utilizing La_2O_3 in optimal conditions. The reaction yield, time, and melting points of the synthesized products are shown in **Table 2**. According to the results in this table, all aryl aldehydes produced the corresponding 2-amino-4-aryl-4*H*-pyrans in short reaction times with good yields (**Table 2**, entries 1a-14a). Therefore, La_2O_3 was a very effectual and general catalyst for the synthesis of this class of heterocyclic compounds.

The Lanthanum (III) cation of La_2O_3 is a Lewis acid, and the oxide anion is a base; thus, it can be used as a both acidic and basic catalyst in the reaction, i.e. as a double-functional catalyst. According to the dual-functionality of La_2O_3, we suggested a plausible mechanism for the production of 2-amino-4-aryl-4*H*-pyrans (**Scheme 2**); this mechanism is consistent with previous literature [21, 23, 25]. Firstly, malononitrile is transformed from intermediate I to intermediate **I** *via* attracting a proton by the basic site of La_2O_3 and then giving back the proton.



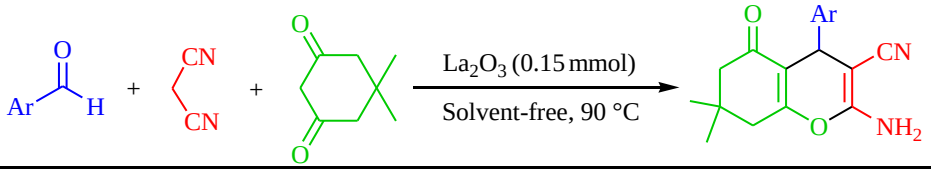
Scheme 1 The model reaction for the preparation of 2-amino-4-aryl-4*H*-pyrans

Table 1 Influence of various parameters on the model reaction.

Entry	The catalyst (mmol)	Temp. (ºC)	Time (min)	Yield[a] (%)
1	0.10	90	35	84
2	0.15	90	10	93
3	0.17	90	10	93
4	0.15	85	15	86
5	0.15	95	10	93
6	-	120	60	31

[a]Isolated yield.

Table 2 Synthesis of 2-amino-4-aryl-4*H*-pyrans catalyzed by La_2O_3

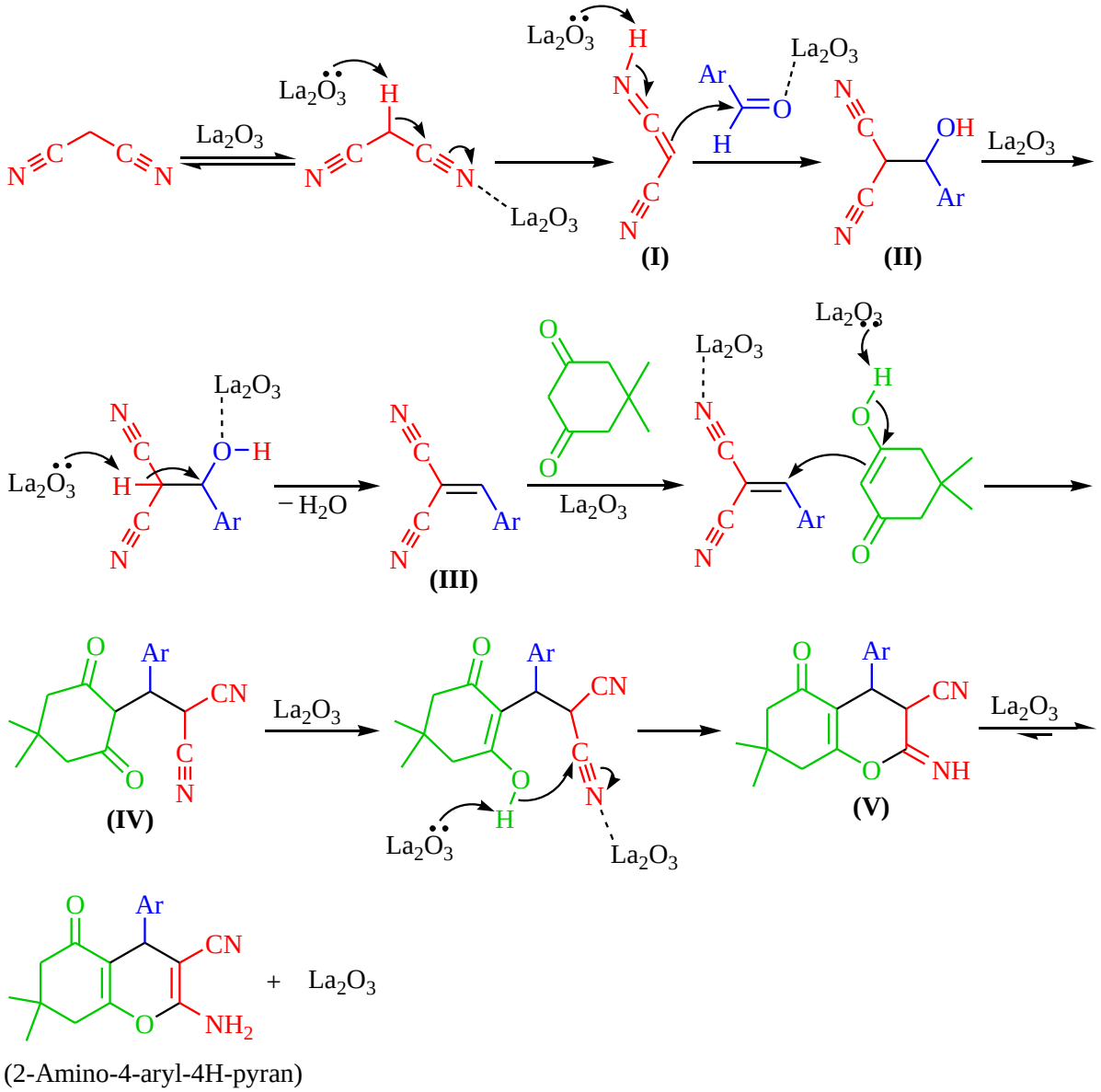


Product	Ar	Time (min)	Yield (%)[a]	M.p. (ºC) Found (Reported)
1a	C_6H_5	10	87	230-232 (232-235) [17]
2a	2-$O_2NC_6H_4$	15	91	183-185 (181-183) [19]
3a	3-$O_2NC_6H_4$	20	83	208-210 (206-208) [17]
4a	4-$O_2NC_6H_4$	20	87	178-180 (181-183) [25]
5a	2-ClC_6H_4	20	80	208-210 (210-212) [19]
6a	3-ClC_6H_4	10	88	226-228 (228-229) [21]
7a	4-ClC_6H_4	10	93	209-211 (212-213) [18]
8a	2,4-$Cl_2C_6H_3$	10	92	181-183 (181-183) [25]
9a	4-FC_6H_4	20	89	195-197 (196-198) [25]
10a	2-BrC_6H_4	15	83	159-161 (160-161) [25]
11a	3-$MeOC_6H_4$	10	92	193-195 (195-197) [18]
12a	4-$MeOC_6H_4$	20	87	196-198 (194-198) [19]
13a	2,4-$Me_2C_6H_3$	20	81	111-112
14a	4-HOC_6H_4	10	90	218-220 (216-218) [21]

[a]Isolated yield.

Nucleophilic attack of **I** (*via* helping the catalyst) to the activated aldehyde (by Lewis acidic site of La_2O_3) affords **II**. Removing a H_2O molecule from **II**, by basic and acidic sites of the catalyst, gives **III**. Afterward, **IV** is formed by Michael-type addition of enol-form of dimedone to intermediate **III**; this step is also catalyzed by lanthanum (III) oxide. Intramolecular addition of the OH group to the activated nitrile by the catalyst forms **V**. Finally, the product is produced by tautomerization of **V** using La_2O_3.

To indicate the ascendancy of La_2O_3 with other catalysts used in the literature in the preparation of 2-amino-4-aryl-4*H*-pyrans, this bifunctional catalyst was compared in terms of the reaction time, yield, type of solvent, and temperature with the following catalysts; the outcomes are demonstrated in **Table 3**. As it is clear from the Table, our catalyst is better for the following reasons: (i) it is better in terms of reaction times, yields, and/or reaction temperature, (ii) La_2O_3 is commercially available; however, most of the catalysts aren't commercially available, and must be synthesized (this topic increases the time and energy consumption, cost and environmental pollution), (iii) performing the synthesis in solvent-free conditions, and (iv) in our procedure, there is no need to additional energy source, e.g. ultrasound irradiation.



Scheme 2 Reaction mechanism for the construction of 2-amino-4-aryl-4*H*-pyrans.

Table 3 Comparing the efficacy of La_2O_3 with different catalysts mentioned in previous literature for the synthesis of 2-amino-4-aryl-4*H*-pyrans

Catalyst	Conditions	Time Range	Yield range	Ref.
La_2O_3	Solvent-free, 90 ºC	10-20	80-93	This work
$Ca_{9.5}Mg_{0.5}(PO_4)_{5.5}(SiO_4)_{0.5}F_{1.5}$	EtOH/H_2O, 60 ºC	50-150	75-92	[16]
Nanoporous polymers of $Zn_4O(H_2N\text{-}TA)_3$	Solvent-free, 60 ºC	270-360	85-95	[17]
MNP@SiO_2-imid-PMA^n	H_2O, r.t., ultrasound	4-14	89-96	[18]
MNP@SiO_2-imid-PMA^n	H_2O, reflux	5-50	82-96	[18]
SiW_{11}Fe-APSCMNPs	Solvent-free, 80 ºC	20	62-93	[19]
Fe_3O_4@p(2‑VP)IL	H_2O, reflux	20-30	80-98	[20]
Phenylboronic acid	EtOH/H_2O, reflux	30	61-88	[21]
$NH_4(H_2PO_4)_2$	EtOH, reflux	40-80	84-93	[22]
[cmmim]Br	Solvent-free, 110 ºC	1-30	84-97	[23]
Thiamine mononitrate	H_2O/EtOH, ultrasound, 40 ºC	20-60	80-94	[24]
Nano-Fe_3O_4@SiO_2@R-NMe_2	EtOH, reflux	90-240	86-98	[25]

3.2. The preparation of pyrazolopyranopyrimidines by La_2O_3

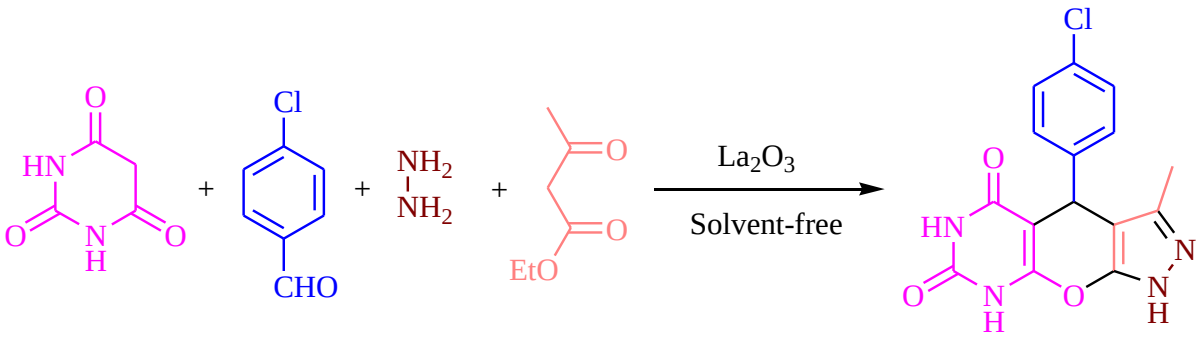
The catalytic ability of La_2O_3 in the absence of solvent was studied for the synthesis of pyrazolopyranopyrimidines after the successful production of 2-amino-4-aryl-4*H*-pyrans using it. For this purpose, after selecting the model reaction among ethyl acetoacetate (1 mmol), hydrazine hydrate (1 mmol), barbituric acid (1 mmol), and 4-chlorobenzaldehyde (1 mmol), initially, the progress of the reaction was monitored in the absence of a catalyst during 60 min at 120 ºC, which was associated with a very low yield of 38% (**Table 4**, entry 1). Then, the influence of temperature and amount of catalyst was evaluated in the range of 50- 70 °C with values of 0.10, 0.15, and 0.20 mmol of La_2O_3 (**Table 4**, entries 2-6). Examining the results showed that the use of 0.15 mmol La_2O_3 at 60 °C is the best condition for the progress of the reaction in the absence of solvent (**Table 4**, entry 3).

To pinpoint the generality and efficacy of La_2O_3, different aryl aldehydes were reacted with ethyl acetoacetate, hydrazine hydrate and barbituric acid under optimal conditions (**Table 5,** entries 1b-13b). As the results of **Table 5** show, since all aryl aldehydes, including benzaldehyde, and aryl aldehydes substituted with electron acceptor or donor atoms or groups, produce the relevant products in a short time and with high efficiency; therefore, La_2O_3 is a powerful and general catalyst for this reaction.

As illustrated in the reaction mechanism (**Scheme 3**), La_2O_3 catalyzes the reaction through the activation of electrophiles and nucleophiles as well as helping tautomerization and elimination of H_2O [28, 31].

One of the most important factors regarding heterogeneous catalysts from an economic and environmental point of view is the ability to recycle and reuse catalysts. Therefore, the reusability of La_2O_3 in the synthesis of derivative **7a** was studied and the results were summarized in **Fig. 1**. According to the obtained results, a very slight decrease in the activity of our bifunctional catalyst was seen after 6 runs.

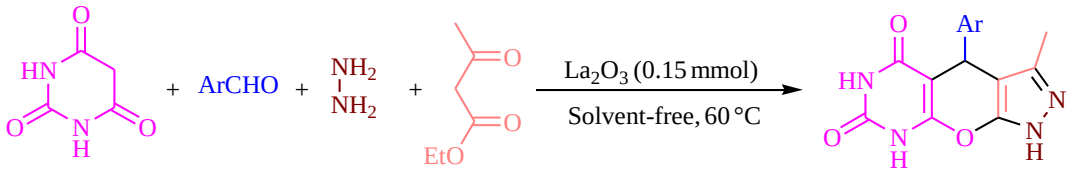
Table 4 Influence of various parameters on the model reaction



Entry	The catalyst (mmol)	Temp. (°C)	Time (min)	Yield[a] (%)
1	-	120	60	38
2	0.10	60	50	78
3	0.15	60	15	97
4	0.20	60	15	97
5	0.15	50	25	83
6	0.15	70	15	97

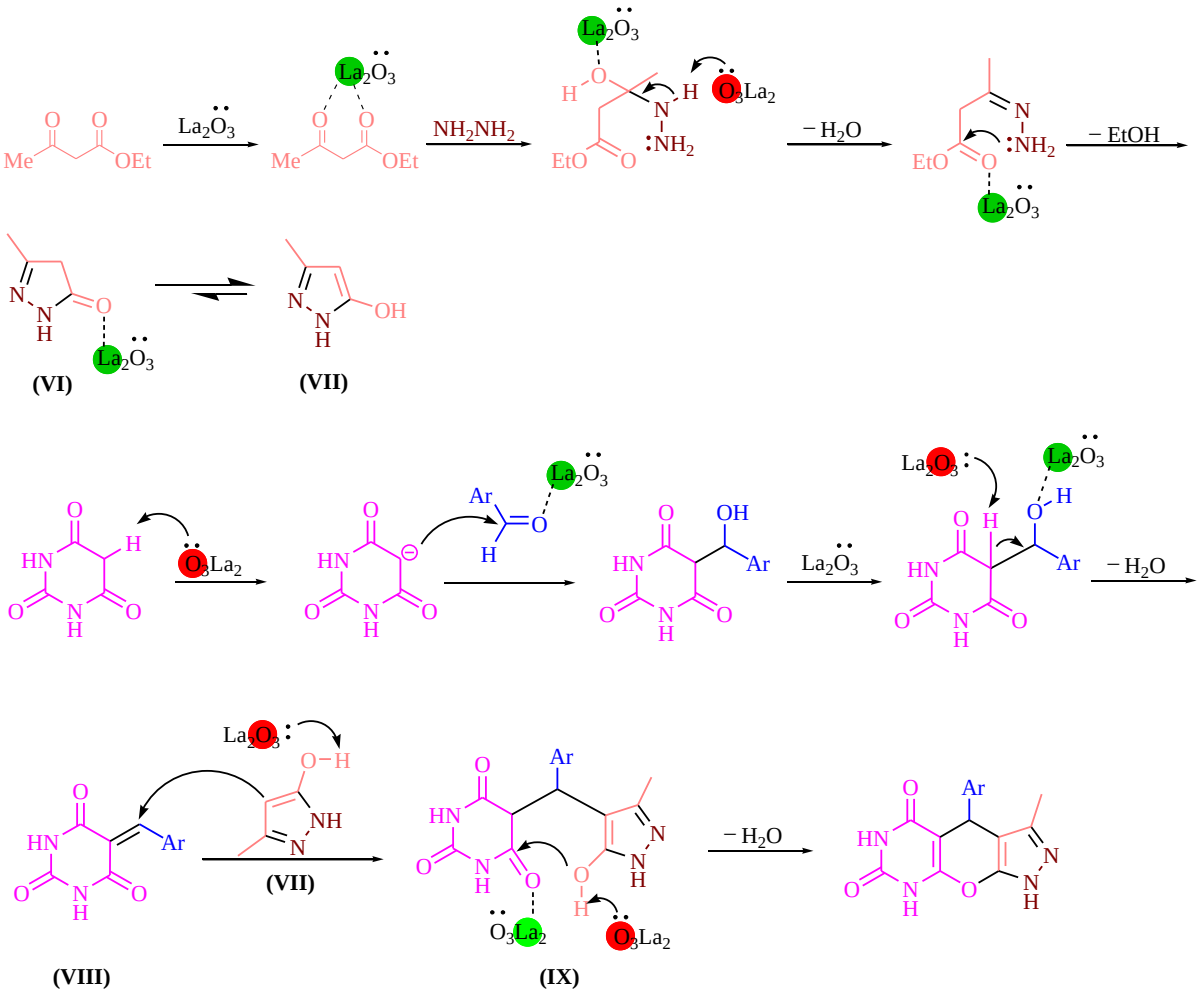
a: Isolated Yields

Table 5 Synthesis of pyrazolopyranopyrimidines usingLa_2O_3



Product	Ar	Time (min)	Yield (%)[a]	M.p. (°C) Found (Reported)
1b	C_6H_5	15	92	217-219 (218-219) [30]
2b	2-$O_2NC_6H_4$	10	98	208-210 (208-209) [30]
3b	3-$O_2NC_6H_4$	13	97	265-267 (266-267) [30]
4b	4-$O_2NC_6H_4$	10	98	233-235(233-234) [30]
5b	2-ClC_6H_4	15	93	222-224 (223-225) [30]
6b	3-ClC_6H_4	16	91	245-247 (246-247) [30]
7b	4-ClC_6H_4	15	97	221-223 (222-223) [30]
8b	2,4-$Cl_2C_6H_3$	22	90	232-234 (233-234) [30]
9b	4-FC_6H_4	12	93	237-239 (237-238) [30]
10b	2-BrC_6H_4	21	92	249-251 (250-251) [30]
11b	3-$MeOC_6H_4$	20	90	221-223 (221-222) [30]
12b	4-$MeOC_6H_4$	20	91	229-231 (230–231) [34]
13b	4-HOC_6H_4	20	92	261-223 (260-262) [34]

[a]Isolated yield.



Scheme 3 Reaction mechanism for the synthesis of pyrazolopyranopyrimidines

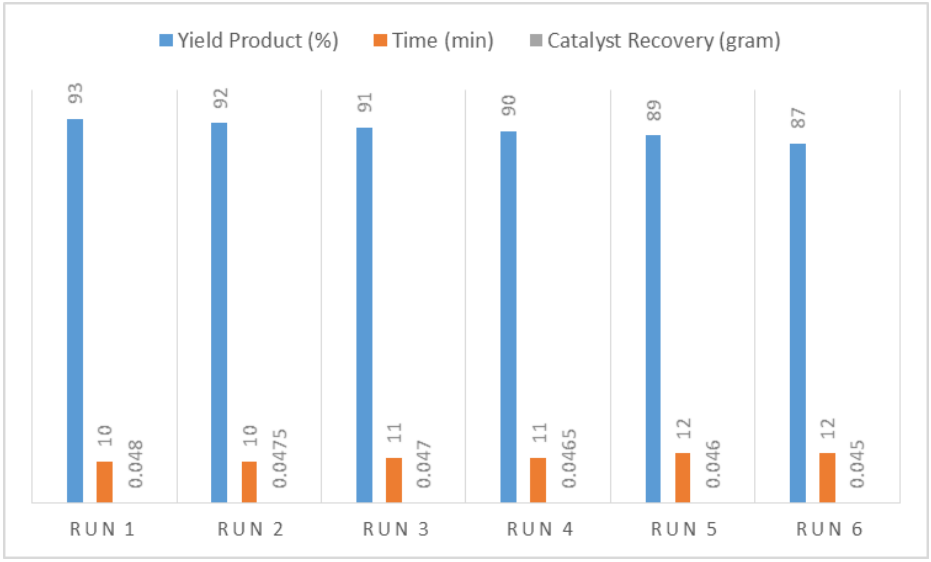


Fig. 1 Reusability of La_2O_3 in the preparation of derivative **7a**

The activity of La_2O_3 compared with other reported catalysts for the production of 2-Amino-4-aryl-4*H*-pyrans and pyrazolopyranopyrimidines were carefully evaluated. The results are summarized in **Table 6**. Based on these results, La_2O_3 has a high activity, because it was superior to most catalysts in at least three of the factors compared in this table (time, yield, temperature, and reaction medium).

Table 6 Comparing of the effectiveness of La_2O_3 with different catalysts mentioned in previous literature for the synthesis of pyrazolopyranopyrimidines

Catalyst	Conditions	Time range (min)	Yield range (%)	Ref.
La_2O_3	Solvent-free, 60 ºC	10-22	90-98	This work
β-Cyclodextrin	H_2O, 50 ºC., ultrasound	25–70	84–93	[26]
TiO_2 NWs	EtOH/H_2O, reflux	45–100	83–95	[27]
ChCl:Urea	EtOH, 80 ºC	60	75–92	[28]
Cu^{2+}@MSNs-$(CO_2^-)_2$	H_2O, rt	60–120	75–92	[29]
Meglumine	H_2O, rt	15–360	83–95	[30]
[MerDABCO-SO_3H]	H_2O, 80 ºC	3-25	73-94	[31]
F-HNTs	H_2O, reflux	30–50	90-96	[32]
OMWCNTs	EtOH/H_2O, reflux	60-100	85-94	[33]
Starch@Fe_3O_4	H_2O, rt, ultrasound	10-25	85-98	[34]
Oleic acid[I]	EtOH, rt	720	84	[8]
Oleic acid[II]	EtOH, reflux	15	78-87	[8]

[I] In this method, only the benzaldehyde derivative was synthesized through a multi-component reaction.
[II] In this method, pyrazolopyranopyrimidine derivatives were synthesized through a domino reaction.

4. Conclusions

Concisely, we have introduced an extremely effective, general, recyclable, and reusable dual-functional catalyst that shows remarkable catalytic activity in one-pot multicomponent preparation of 2-amino-4-aryl-4*H*-pyrans and pyrazolopyranopyrimidines in the absence of solvent. Our synthetic routes were simple, mild, and efficient and the reaction products were isolated by easy crystallization procedure and do not need any further purification steps. Other encouraging aspects in both presented syntheses include providing products in short times with high yields, simplicity, the use of a commercially available heterogeneous catalyst with recyclability and reusability up to 6 runs without significant reduction in activity, and good compliance with green chemistry principles.

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Conflicts of interest

The authors declare that they have no conflict of interest.

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