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#### Short communication

# Tin and copper species dispersed on a metal-organic framework as a new catalyst in aerobic Baeyer-Villiger oxidation: An insight into the mechanism



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# A R T I C L E I N F O A B S T R A C T Keywords: Metal-organic frameworks (MOF) containing tin and copper metal salts were used as catalysts in the Baeyer-Villiger oxidation of cyclohexanone. The outcome was a synergistic effect on the catalyst's efficiency, which resulted from the simultaneous presence of copper and tin species in the MOF. The catalyst with high metal content showed the intermolecular dehydration of benzoic acid and the formation of benzoic anhydride in a side reaction as well as the Baeyer-Villiger oxidation reaction with a decline in efficiency. The optimized catalyst promoted the Baeyer-Villiger reaction in a high yield without the formation of benzoic anhydride.

#### 1. Introduction

The Baeyer-Villiger (B-V) oxidation reaction is one of the most important processes in the field of organic transformation chemistry. In this reaction, ketones become lactones or esters, which are valuable intermediates in a wide range of chemical industries [1,2]. E-Caprolactone, which is a chemical produced in the B-V reaction, plays a major role in the pharmaceutical and medical fields as a monomer in the preparation of biodegradable poly-ε-caprolactone [3-5]. In this context, the evolution of improved productive and selective catalysts in the B-V oxidative reaction has attracted the attention of many scientists working in the catalyst field. Consequently, several catalysts, including mesoporous silica [6], Cu-Fe<sub>3</sub>O<sub>4</sub> supported mesoporous silica [7], Keggin-type heteropoly salts [8], graphite [9], and carbon nanotubes [10] have been introduced. However, these catalysts have some limitations and drawbacks, including high metal loading, prolonged reaction time, relatively low activity, and difficult synthesis. Another issue in some previously reported catalyst systems is the necessary use of expensive oxidizers or environmentally harmful oxidizers, such as hydrogen peroxide and peracids. The use of hydrogen peroxide results in the formation of water as a byproduct in the reaction medium, which leads to the hydrolysis of the product. To overcome this problem, hydrogen peroxide was replaced by oxygen or air in the presence of aldehyde as a sacrificing agent, which is known as the Mukaiyama method. However, this method is problematic because it requires the use of pure oxygen and benzaldehyde greater than the stoichiometric ratio. Earlier studies showed the beneficial effects of deposited tin and copper species, as the Lewis acid sites on various supports, including silica [11], zeolites [12,13],  $TiO_2$  [14], and graphene oxide [15], in an oxidation reaction. Among the works reported so far, only Huo et al. used tin and copper simultaneously in the B-V reaction [11]. The aim of the present study was to create an outstanding catalyst by integrating copper and tin species in porous metal-organic framework (MOF), which are potentially powerful platforms in catalysis [16–19], and to take advantage of both species and the MOF simultaneously. A further aim was to make this reaction economical and environmentally friendly by designing a catalyst that is less cost- and time-consuming to synthesize, decreasing the amount of metal loading and using less benzaldehyde as the sacrificing agent.

Metal-organic frameworks are hybrid porous materials made of organic linkers and metal clusters or ions that are suitable for stabilizing metal nanoparticles [20]. In addition, the size of the metal particles can be adjusted by the cavities and windows of the MOF. The control of the metal particle size allows for the access to active catalytic sites and overcomes several obstacles, such as the hard recovery from the reaction environment, agglomeration, and the accumulation of nanoparticles [21]. Among the various MOFs previously reported regarding the loading of metal nanoparticles and metal ions, MIL-101(Cr) has attracted particular attention because of its stability and high resistance to hydrolysis [22,23].

Despite all conditions provided for the intermolecular dehydration of benzoic acid in previous studies of the B-V reaction in the presence of  $Cu^{II}$  and  $Sn^{IV}$  under air/O<sub>2</sub> [24,25], the formation of benzoic anhydride was not observed. However, the results of this work showed that the benzoic anhydride is produced by the intermolecular dehydration of benzoic acid in the presence of  $Cu^{II}$  and  $Sn^{IV}$  under air as an oxidant,

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and the formation of benzoic anhydride could be controlled and eliminated by selecting the optimal metal loading. Therefore, it was deemed beneficial to investigate the effects of metal species loading on the conversion of benzaldehyde as the sacrificing agent to benzoic anhydride by the dehydration reaction among the benzoic acid molecules produced by benzaldehyde oxidation. In this study, MIL-101(Cr) was used as a support to fabricate a potent catalyst. To the best of our knowledge, this is the first study to use an MOF to prepare heterogeneous catalysts for the B-V oxidation reaction. In addition, to obtain further insights and clarify the reaction mechanism, the effects of metal loading were investigated. The results showed that by using the present catalyst, the amount of benzaldehyde in the reaction decreased by 25% compared with previous studies. These results indicate that the remarkable performance of this catalyst in the critical cyclohexanone oxidation reaction and the production of  $\varepsilon$ -caprolactone could open a new direction in the use of different MOFs to promote the B-V reaction and introduce a new and reliable alternative that would reduce both the cost and the environmental risks associated with this reaction.

#### 2. Experimental

#### 2.1. Materials and instrumentation

All the chemicals and solvents used in this study were acquired from Merck, and they were applied as received. The infrared spectra were obtained on a Bomem MB-Series FTIR spectrometer. The powder X-ray diffraction (XRD) measurements were performed at room temperature by a STOE diffractometer using monochromatized Cu  $K_{\alpha}$  radiation ( $\lambda = 0.15418$  nm). The nitrogen physisorption technique was applied to measure the surface area and textural properties of the prepared catalysts at -196 °C using a Micromeritics Tristar II 3020 apparatus. The scanning electron micrographs (SEM) were obtained in the secondary electron contrast using a MIRA3 TESCAN field-emission instrument. The same instrument was used to carry out the energy-dispersive X-ray (EDX) analyses. The concentrations of tin and copper were measured using an Espectro Arcos (AMETEK) inductively coupled plasma optical emission spectrometer (ICP-OES). The conversion of the reaction was estimated by an Agilent 7890 A (G3440A) GC. The byproducts were identified by GC-MS (TRACE MS/ ThermoQuest-Finnigan) with a DB-5 column.

#### 2.2. Synthesis of MIL-101(Cr) and Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr)

The synthesis of MIL-101(Cr) was carried out following the previously reported method [26]. To synthesize Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr) with different percentages of metal, first different amounts of copper nitrate and tin chloride were completely dissolved in 5 mL ethanol and added to 0.5 g of MIL-101(Cr) that had been pre-activated at 160 °C. The resulting mixture was stirred for 24 h, and then heated to 50 °C until complete dryness of the solvent (Fig. 1). The prepared catalysts with different percentages 0.4, 0.8; 0.6, 1.2; 0.8, 1.6; 0.8, 4.8; 2.4, 1.6; and 2.4, 4.8 wt% of copper and tin ions were designated as Cu<sup>II</sup>-Sn<sup>IV</sup>@ MIL-101(Cr)/A, B, C, D, E, and F, respectively, in Table 1.

### 2.3. General procedure for catalytic reactions and the regeneration of catalyst

The aerobic B-V oxidation of cyclohexanone was performed in a flask equipped with a refluxing condenser. Cyclohexanone (0.196 g, 2 mmol) and benzaldehyde (0.318 g, 3 mmol) were poured into a flask containing 5 mL of dichloroethane (DCE), and the reaction was carried out at 50 °C for 5 h in the presence of 50 mg of the activated catalyst while air was passed continuously through the reaction mixture. After each catalytic cycle, Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr)/C was recovered by centrifugation, and after washing with dichloromethane, it was dried under vacuum at 150 °C and then used in the subsequent step of the catalytic test. The reaction conversion and the identification of side products were performed by GC and GC–MS analyses, respectively.

#### 2.4. Leaching test

After 3 h of reaction, the catalyst was separated from the hot reaction matrix. The reaction then was continued for 6 h with the filtrate in the absence of the catalyst. After the catalyst was removed, the yield of the reaction product did not increase. This result indicated that the catalytically active sites, which were  $Cu^{II}$  and  $Sn^{IV}$  species, remained intact in the catalyst and did not leach into the reaction solution.

#### 3. Results and discussion

#### 3.1. Characterization of catalysts

Various analyses were performed to identify the structure and morphology of the prepared catalysts. Compared with the FTIR spectrum of MIL-101(Cr), the FTIR spectrum of Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr) did not show new absorption bands. For this reason, ICP and EDX analyses were carried out to establish the presence of metal salts in the MOF.

The XRD patterns of the catalysts that were prepared using MIL-101(Cr) as the support are illustrated in Fig. 2. The XRD results confirmed that the prepared catalysts possessed crystalline structures expected in the relevant MOF, and the metal loading on them had no effect on their crystalline structures. Because of the low amounts of loaded metal species and their uniform distribution on MIL-101(Cr), reflections of the loaded metal salts did not emerge in the Cu<sup>II</sup>-Sn<sup>IV</sup>@ MIL-101(Cr) diffraction pattern.

The representative nitrogen adsorption-desorption isotherms of the Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr) catalyst and the bare support are shown in Fig. S1 (Electronic Supplementary Information, ESI). The BET surface area of 2214 m<sup>2</sup>g<sup>-1</sup> and pore volume of 1.13 cm<sup>3</sup>g<sup>-1</sup> was determined for the synthesized MIL-101(Cr). After impregnation with different amounts of copper and tin salts in MIL-101(Cr), the BET surface area and total pore volume were decreased to 1943 m<sup>2</sup>g<sup>-1</sup> and 0.99 cm<sup>3</sup>g<sup>-1</sup> for Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr)/C and 1713 m<sup>2</sup>g<sup>-1</sup> and 0.88 cm<sup>3</sup>g<sup>-1</sup> for Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr)/F. These results confirmed the successful loading of metal salts inside the cavities.

Fig. 3 (insert a) shows the SEM image of the  $Cu^{II}$ -Sn<sup>IV</sup>@MIL-101(Cr)/C catalyst. As the micrograph shows, the octahedron morphology of MIL-101(Cr) was preserved after the metal salts were loaded. The elemental composition of the  $Cu^{II}$ -Sn<sup>IV</sup>@MIL-101(Cr) samples was determined by EDX analysis, and the Cu and Sn loadings





#### Table 1

The results of the cyclohexanone	oxidation catalyzed b	v various catalysts	under air atmosphere at 50 °C.
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Entry	Catalyst	Metal loading (%) Cu-Sn	Yield of $\epsilon$ -caprolactone (%)	Yield of benzoic acid (%)	Yield of benzoic anhydride (%)	TON <sup>b</sup>
1	Blank	-	12	100	-	-
2	MIL-101(Cr)	-	35	100	-	-
3	Cu <sup>II</sup> @MIL-101(Cr)	1.55-0	23	100	-	38
4	Sn <sup>IV</sup> @MIL-101(Cr)	0-1.54	44	100	-	131
5	Cu <sup>II</sup> -Sn <sup>IV</sup> @MIL-101(Cr)/A	0.37-0.79	42	100	-	136
6	Cu <sup>II</sup> -Sn <sup>IV</sup> @MIL-101(Cr)/B	0.59-1.18	72	100	-	151
7	Cu <sup>II</sup> -Sn <sup>IV</sup> @MIL-101(Cr)/C	0.78-1.57	100	100	-	154
8	Cu <sup>II</sup> -Sn <sup>IV</sup> @MIL-101(Cr)/C	1.57-1.56	65	85	5	69
9	Cu <sup>II</sup> -Sn <sup>IV</sup> @MIL-101(Cr)/D	0.78-4.74	60	82	7	46
10	Cu <sup>II</sup> -Sn <sup>IV</sup> @MIL-101(Cr)/E	2.36-1.54	19	70	10	15
11	Cu <sup>II</sup> -Sn <sup>IV</sup> @MIL-101(Cr)/F	2.36-4.71	19	86	14	10
$12^{a}$	Cu <sup>II</sup> -Sn <sup>IV</sup> @MIL-101(Cr)/F	2.36-4.71	0	63	24	-
13	Cu <sup>II</sup> @MIL-101(Cr)	2.30-0	17	73	8	18
14	Sn <sup>IV</sup> @MIL-101(Cr)	0–4.75	86	92	4	86

Reaction conditions: cyclohexanone (2 mmol), benzaldehyde (3 mmol), and catalyst (50 mg) in DCE (5 mL), 50 °C, 5 h, under pumping air (20 mL min<sup>-1</sup>). <sup>a</sup> Without cyclohexanone.

 $^{\rm b}\,$  Turnover number is calculated based on the amount of copper and tin for  $\epsilon\text{-caprolactone.}$ 



Fig. 2. XRD patterns of (a) simulated MIL-101(Cr), (b) synthesized MIL-101(Cr), (c) Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr)/C, and (d) Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr)/F.

were also determined by ICP analysis. As shown in Fig. 3, the results of the EDX analysis revealed that the as-prepared catalyst contained Cr, Cu, Sn, C, O, N, and Cl. The EDX elemental mapping (Fig. 3, insert b) results also implied that the Cu and Sn species were homogeneously dispersed on the surface of MIL-101(Cr). The results of the ICP analysis showed that the contents of copper and tin in the Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr)/A, B, C, D, E, and F were 0.37, 0.79; 0.59, 1.18; 0.78, 1.57; 0.78, 4.74; 2.36, 1.54; and 2.35, 4.71 wt%, respectively.

#### 3.2. Evaluation of factors effecting the cyclohexanone oxidation reaction

The aerobic B-V oxidation of cyclohexanone was carried out in the presence of benzaldehyde as a sacrificial agent under various conditions to determine the optimal conditions. A main problem in the B-V reaction under Mukaiyama conditions is the need to use large amounts of benzaldehyde, which results in substantial amounts of benzoic acid in the reaction media. To resolve this issue, different quantities of benzaldehyde in the reaction were examined (Table S1), which revealed that 2:3 mol was the optimal ratio of cyclohexanone to benzaldehyde.

The investigation of temperature as an active parameter on the catalytic reaction demonstrated that by increasing the temperature from 30 °C to 50 °C, a significant increase in the yield of the  $\varepsilon$ -caprolactone (35–100%) occurred, whereas increasing the temperature to 70 °C resulted in a downward trend in the product yield (Table S2). Furthermore, the effect of the catalyst amount on the reaction progression was investigated and results are listed on Table S3. As the results show by increasing the amount of the catalyst to 50 mg, the reaction yield increased, and the further addition of the catalyst did not increase the yield.

The reaction was carried out in the presence of various catalysts to evaluate the role of MIL-101(Cr) and the metal species loaded on it in the cyclohexanone oxidation process. The results are listed in Table 1. Notably, in the absence of the catalyst, the yield of  $\varepsilon$ -caprolactone was 12%. In addition, as Table 1 shows, the Lewis acid centers present in the MIL-101(Cr) had a positive effect on the cyclohexanone aerobic



Fig. 3. EDX analysis, SEM image (insert a), and elemental mapping (insert b) of Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr)/C.



Fig. 4. Effect of reaction time on the conversion of the B-V oxidation of cyclohexanone with  $Cu^{II}$ -Sn<sup>IV</sup>@MIL-101(Cr)/C as catalyst (a) and leaching test (b).

oxidation reaction [27] because, as can be seen, in the absence of MIL-101(Cr), the reaction efficiency is reduced to 12% (entry 1) from 35% (entry 2). However, copper nitrate by blocking chromium sites through the coordination of nitrate groups, deactivated them and decreased the reaction efficiency (entry 3). The use of copper or tin solely in the catalyst results in low catalyst performance (entries 3 and 4). In contrast, the simultaneous presence of copper and tin species in MIL-101 (Cr) had a synergistic effect on the reaction efficiency (entry 7). It was also observed that by increasing the amount of loaded metal species to a certain value (entry 11), both the efficiency and selectivity of the catalyst in the formation of  $\varepsilon$ -caprolactone were significantly decreased. Instead, benzoic anhydride started to form as a side product in another pathway.

For further insight, a nitrogen adsorption-desorption analysis of the prepared catalysts with different amounts of metal loading (Fig. S1, ESI) was carried out to determine the role of the surface area and the size of the catalyst cavities in the B-V oxidation performance of cyclohexanone. The outcomes of these analyses revealed that the decreases in the surface area and the size of the cavities in catalysts with higher metal species loadings were not significant enough to contribute to the decrease in catalytic activity. The results showed that as the metal percentage increased, the oxidative strength of the catalyst increased, and part of the benzaldehyde was oxidized to benzoic acid [28] before entering into the main reaction pathway and the production of  $\epsilon$ -caprolactone. Finally, by decreasing the amount of benzaldehyde in the reaction medium as a sacrificial agent in the B-V reaction, the yield of εcaprolactone was declined. In contrast, large amounts of benzoic acid were produced in the reaction medium, which was converted to benzoic anhydride during the intermolecular dehydration process [29-31]. It should be noted that according to all previous reports of the B-V oxidation reaction in the presence of benzaldehyde as a sacrificial agent, the formation of benzoic acid as a byproduct was inevitable [9-11,24,25,32,33]. However, the formation of benzoic anhydride during the B-V oxidation reaction was observed for the first time in this study. The formation of these byproducts was confirmed by GC-MS analysis. As shown in Table 1, by increasing the metal loading above a certain amount (entry 11), conditions were provided for the intermolecular dehydration reaction, and benzaldehyde was involved in both the B-V oxidation reaction and the intermolecular dehydration reaction, which resulted in a decrease in the  $\varepsilon$ -caprolactone yield (yield of ɛ-caprolactone: 19, benzoic acid: 86 and benzoic anhydride: 14%). Although carboxylic anhydrides are an important class of the highly active organic compounds that are involved as intermediates in organic synthesis, especially in the preparation of drugs and peptides, they were considered a byproduct in this reaction. In addition, the results showed that tin and copper had a synergistic effect on the formation of benzoic anhydride (Table 1, entry 11), and removal of either from the catalyst led to a decrease in benzoic anhydride yield (entries 13 and 14). As Table 1 shows, the limit value of Cu content in catalyst structure for the absence of benzoic anhydride in the reaction mixture was 0.78% (Table 1, entry 7) and by increasing the Cu content to 1.57% (entry 8), 5% benzoic anhydride produced. This limit for Sn is 1.57% (entry 7), and by increasing it to 4.74%, 7% benzoic anhydride produced (entry 9). Notably, by decreasing the metal species loading, particularly the amount of copper, the production of benzoic anhydride was prevented, consequently, leading to the direct reaction to ɛ-caprolactone formation. For further insights into the formation of benzoic anhydride, the reaction was performed in the absence of cyclohexanone with a higher metal loading catalyst (Table 1, entry 12) with respect to the optimized catalyst (entry 7). In the absence of cyclohexanone (entry 12), the competing factor in the use of benzaldehyde was eliminated from the reaction medium. Thus, there was a substantial amount of benzaldehyde in the medium, and it was converted to the benzoic anhydride through intermolecular dehydration of benzoic acid that produced by oxidation of benzaldehyde; consequently, the yield of benzoic anhydride increased from 14 to 24%. To enable the better understanding of the issues discussed above, the proposed mechanism is explained below.

Metal ions as Lewis acid centers play a crucial role in advancing the B-V reaction [11,14,24]. Initially, the coordination of benzaldehyde to metal ions results in the formation of benzoyl radical, PhCO $\cdot$ , and then in the presence of molecular oxygen and benzaldehyde, perbenzoic acid forms. Then, Lewis centers by activating cyclohexanone make it susceptible to nucleophilic attack by the peracid to form the Criegee complex. Subsequently, the Criegee adduct rearrangement results in the formation of  $\varepsilon$ -caprolactone and benzoic acid [11,14,24].

In the present study, the cyclohexanone oxidation reaction was performed at different times to evaluate the effect of time on the reaction. As shown in Fig. 4a, when the reaction time increased from 1 to 5 h, the yield of the product increased from 34% to 100% and then remained constant over longer a duration. A hot filtration test was performed to confirm the heterogeneity of the catalyst (Fig. 4b). As shown, after the removal of the catalyst from the reaction medium, the catalytic process did not progress, which indicated the heterogeneous nature of the catalyst and the absence of leaching in the catalytically active sites.

Table 2

omparative chart of catalytic activity of C	ı"-Sn	<sup>w</sup> @MIL-101(Cr)/C wi	ith other solid catalysts in the B-V	oxidation of cyclohexanone u	sing air/O <sub>2</sub> and benz	zaldehyde.
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Entry	Catalyst (mg)	substrate/aldehyde <sup>a</sup>	T (°C)	Time (h)	Yield (%)	Oxidant	Ref.
1	Ketjen Black (5)	10:20	50	6	91	O <sub>2</sub>	[32]
2	MnALPO-36 (150)	1:3	50	6	76.4	$O_2$	[33]
3	Graphite (20)	4:4	120	3	99	$O_2$	[9]
4	c-MWCNTs (20)	2:4	50	8	> 99	0 <sub>2</sub>	[10]
5	Fe – Cu bimetal oxide (25)	2:5	50	4	> 99	air	[25]
6	Cu-MCM-41 (50)	2:4	50	3	99	O <sub>2</sub>	[24]
7	Fe <sub>3</sub> O <sub>4</sub> -l-dopa-Cu <sup>II</sup> /Sn <sup>IV</sup> @m,mSiO <sub>2</sub> (50)	2:4	50	6	> 99.9	air	[11]
8	Cu <sup>II</sup> -Sn <sup>IV</sup> @MIL-101(Cr) (50)	2:3	50	5	100	air	This work

<sup>a</sup> The mole ratio of cyclohexanone to benzaldehyde.



**Fig. 5.** Reuse of  $Cu^{II}$ -Sn<sup>IV</sup>@MIL-101(Cr)/C for the B-V oxidation using air and benzaldehyde. Reaction conditions: cyclohexanone (2 mmol), benzaldehyde (3 mmol), and catalyst (50 mg) in DCE (5 mL), 50 °C, 5 h, under pumping air (20 mL min<sup>-1</sup>).

Table 2 shows the comparison of the catalytic performance of the prepared catalyst in this work with other catalysts in previous reports of the cyclohexanone oxidation reaction under Mukaiyama conditions. As Table 2 shows, the catalyst introduced in this work had several advantages, including the use of lower amounts of benzaldehyde as the sacrificial agent, lower reaction time, increased facility, and the less expensive synthesis of the catalyst compared with previously reported catalysts.

In the use of catalysts in various processes, a crucial point is their ability to be reused without any considerable reduction in their catalytic performance. Thus, in the present study catalytic recovery experiments were conducted, and the results confirmed that  $Cu^{II}$ -Sn<sup>IV</sup>@ MIL-101(Cr)/C was successful in several recovery cycles without a noticeable decrease in efficiency (Fig. 5). The results showed that, no decrease in the product yield during the first and second cycles of the catalytic reaction, but in the third cycle, there was a slight decrease in the catalyst performance. To determine the reason for the decline in yield, after the catalyst was applied twice in the reaction, the amount of metal leaching was determined by ICP. The results indicated the 10% leaching of both copper and tin metals, which resulted in a decrease in the reaction efficiency in the third cycle.

#### 4. Conclusion

 $Cu^{II}$ -Sn<sup>IV</sup>@MIL-101(Cr) catalysts were prepared with various loadings of copper and tin species and evaluated in the aerobic oxidation of cyclohexanone. The results showed that the simultaneous presence of copper and tin species in the catalyst had a synergistic effect on catalytic efficiency and selectivity. In addition, the results of this study showed that the amount of metal loading played an important role in directing the reaction to the desired product, which was  $\varepsilon$ -caprolactone. Furthermore, increasing the metal loading resulted in side reactions and benzoic anhydride formation through the intermolecular dehydration of benzoic acid, thereby in reducing the main product yield. Moreover, by using Cu<sup>II</sup>-Sn<sup>IV</sup>@MIL-101(Cr)/C as a catalyst, it was possible to decrease the consumption of benzaldehyde by 25%, therefore reducing both the environmental risks associated with the production of benzoic acid and the cost of the B-V reaction.

#### **Declaration of Competing 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.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.catcom.2020.105985.

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