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# **One-pot synthesis of 1-butylpyrrolidine and its derivatives from** aqueous ammonia and 1,4-butandiol over CuNiPd/ZSM-5 catalyst<sup>+</sup>

employed raw materials are readily available and easy to

handle<sup>13-15</sup>. In our recent work, the synthesis of 1-

methylpyrrolidine with >90% yield from BDO and

methylamine via a one-pot method was realized successfully<sup>16</sup>.

As well known, aqueous ammonia is a more attractive nitrogen

source due to its advantages in safety and handling<sup>17, 18</sup>. In the

previous report<sup>19</sup>, 1-BP was obtained from the reaction

between BDO and NH3 over iron catalyst under hydrogen

atmosphere (Scheme 1), and the highest yield of 1-BP was only

38%. It should be noted that 1-BP was not the object of study

in the mentioned reference, which just was one of the products.

The design of catalyst is central to promote 1-BP yields in this route. As is well known, Cu and Ni based catalysts were very

efficient for the N-alkylation of amines or ammonia with alcohols 20. However, it was found that some 1-BePs were

generated using our recently developed Cu and Ni modified

ZSM-5<sup>16</sup>, which derived from the dehydration of BDO moity <sup>21</sup>. 1-BePs are also value-added products, which would be our

next work for synthesis of 1-BePs and this work focuses on 1-

BP. It is an alternative strategy to prompt the yield of 1-BP via

hydrogenation of 1-BePs over a hydrogenation catalyst. It was reported that Pd based catalysts could facilitate both N-alkylation and hydrogenation<sup>22, 23</sup>. Based on the above, a

ternary catalyst (including Pd, Cu and Ni species) supported on

ZSM-5 was further developed, and the catalyst was prepared

by impregnation method (See in SI), which displayed fine

catalytic performance with just low content of Pd (0.2%) for

The optimization of reaction conditions was carried out

carefully, and 76% yield of 1-BP was obtained. There is still 5%

selectivity of 1-BePs in the products, whereas 1-BePs could

convert to 1-BP via further hydrogenation from the view of

practical application. Meanwhile, the relationship between

structure and performance of the prepared catalyst was

preliminarily explored by various characterizations. Therefore,

a simple method for synthesis of 1-BP with good yield was

realized from BDO and NH<sub>3</sub> over developed ZSM-5 catalyst in

this work, in which the employed raw materials were much

cheaper than the routes involving pyrrolidine, and the un-green

reagents or by-products were also avoided. Considering the

simple and economical features in this protocol, it has a

promising perspective for industrial application. Besides, it may provide an alternative method for synthesis of N-

heterocycle derivatives using NH<sub>3</sub> as the nitrogen source.

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synthesis of 1-BP.

The synthesis of 1-butylpyrrolidine and its derivatives (1butylpyrrolidine with a little of 1-butenylpyrrolidines) was developed via one-pot method from ammonia and 1,4-butandiol. Here, the product of 1-butylpyrrolidine was emphatically investigated, and the yield was 76% under optimization conditions. Such route was realized through successive Nalkylation using aqueous ammonia as the nitrogen source over CuNiPd/ZSM-5 catalyst, which was prepared by simple incipient wetness method. In this route, 1,4-butandiol not only participated in the formation of N-heterocycle, but also acted as alkylating reagent. This work offers a straightforward, economical route for 1-butylpyrrolidine and its derivatives.

The development of new reaction route for chemicals synthesis, with more simple, green, and atomic economy features, is always a central theme in chemistry, and catalysis would play an important role in this respect. 1-Butylpyrrolidine (1-BP) and its derivatives (mainly be 1-butenylpyrrolidines, 1-BePs), could be used as organic intermediates<sup>1,2</sup>, which also are important raw materials for the preparation of pyrrolidinium ionic liquids <sup>3-5</sup>. Currently, the traditional synthetic methods for 1-BP and its derivatives mainly include the following processes: 1) the butylation of pyrrolidine with halohydrocarbons, butyraldehyde or 1-butanol 1,6-8, 2) the cyclization of 1,4-dichlorobutane or 1,4-butandiol (BDO) with 1-butylamine <sup>6, 9, 10</sup>. However, the above-mentioned routes often suffer from the high cost of raw materials or environmental pollution (the previous reported routes for the synthesis of 1-BP were listed in Table S1). Firstly, both pyrrolidine and 1-butylamine are relatively expensive raw materials; secondly, using toxic butyraldehyde as the butylation reagent is a hazardous threat to the environment and human health; besides, as butyl halides are involved in the process, it needs a large amount of NaOH to neutralize the inorganic acid by-product (HCl or HBr), which would produce a great deal of waste salts. Herein, we try to develop a simple and economical route for the synthesis of 1-BP and its derivatives through a forthright reaction between NH<sub>3</sub> and BDO, and the product 1-BP was emphatically studied.

Ammoniation and cyclization of BDO with amines to pyrrolidine derivatives with only water by-product could be considered as an atom-efficient and green route<sup>11, 12</sup>, and the

† Electronic Supplementary Information (ESI) available: [details of any 59 supplementary information available should be included here]. See

Catalyst `OH + NH<sub>3</sub> —

Scheme 1 Synthesis of 1-BP from BDO and NH<sub>3</sub>

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### Journal Name

COMMUNICATION

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Initially, the catalytic performance of different catalysts was investigated for the synthesis of 1-BP from BDO and NH<sub>3</sub>, and the results were listed in Table 1. It could be seen that only 8% BDO conversion was obtained in the absence of catalyst (Entry 1, Table 1), and the support HZSM-5 zeolite also showed poor catalytic performance with 32% BDO conversion and 2% 1-BP selectivity (Entry 2, Table 1). As Cu/ZSM-5 and Ni/ZSM-5 were used, although the conversion of BDO increased by a large margin, the selectivity of 1-BP was moderate (22% and 43%, respectively; Entry 3 and 4, Table 1), and 1-BePs including 1-(3-buten-1-yl)-pyrrolidine (1-3-BeP) and 1-(2-buten-1-yl)-pyrrolidine (1-2-BeP) were also formed. While, it was found that full conversion of BDO was achieved and the selectivity of 1-BP increased to 55% with the decline of 1-BePs, when both Cu and Ni were modified on ZSM-5 (Entry 5, Table 1), which implied that the synergistic effect between Cu and Ni species on ZSM-5 improved the N-alkylation of ammonia with BDO and enhanced the hydrogenation of 1-BePs to some extent. Further, the 1-BP selectivity could be up to 76% maintaining the full conversion of BDO as 0.2wt% Pd was added (Entry 6, Table 1). It could be seen that 1-BePs declined distinctly, and

**Table 1** Catalytic performance of different catalysts <sup>a</sup>

the selectivity of 1-3-BeP and 1-2-BeP was 4% and 1%, respectively, which meant that Pdol:species/ppromoted hydrogenation. Meanwhile, the total amounts N-substituted pyrrolidine compounds (including 1-BP and 1-BePs) also increased, which might imply that the introduction of Pd was also benefit to the N-alkylation. Next, the effect of Pd loading was studied. With the increase of the loading of Pd, although the content of 1-BePs reduced, the selectivity of 1-BP declined slightly; while 1-propylpyrrolidine (1-PP) and 1butanol were raised as the by-products (Entry 7-8, Table 1). The reason might be due to the catalytic action of excessive Pd species for the hydrodealkylation of BDO to 1-propanol (which would react with pyrrolidine to 1-PP) and partial hydrogenation of BDO to 1-butanol <sup>24, 25</sup>. Other zeolites as carriers were also screened, such as Y or Beta zeolite (Entry 9-10, Table 1), but the catalytic performance was inferior to ZSM-5 zeolite. Besides, ZSM-5 zeolites with different ratios of Si/Al were tested (entries 11-12, Table 1). It could be seen that, as the ratio of Si/Al was 25 and 300, the yield of 1-BP was 69% and 71 %, respectively, which may mean that ZSM-5 with an 80 Si/Al ratio provided suitable acidity for this process <sup>14</sup>.

Entry	Catalyst <sup>b</sup>	Conv./%	Selectivity/%						
			1-BP	1-PP	THF	1-Butanol	1-3-BeP <sup>c</sup>	1-2-BeP <sup>c</sup>	Others <sup>d</sup>
1	none	7	1		99				
2	H-ZSM-5	32	2		98				
3	3Cu/ZSM-5	69	22		15		26	9	28
4	3Ni/ZSM-5	97	43		7		15	8	27
5	3Cu-3Ni/ZSM-5	100	55		4		13	4	24
6	3Cu-3Ni-0.2Pd/ZSM-5	100	76	3	4	4	4	1	8
7	3Cu-3Ni-0.5Pd/ZSM-5	100	68	11	2	7	2		10
8	3Cu-3Ni-1Pd/ZSM-5	100	72	10	1	14	2		1
9	3Cu-3Ni-0.2Pd/USY	100	50	1	7	10	20	6	6
10	3Cu-3Ni-0.2Pd/Beta	100	62	4	2	2	15	4	11
$11^{e}$	3Cu-3Ni-0.2Pd/ZSM-5	100	69	7	3	1	4	2	14
$12^{e}$	3Cu-3Ni-0.2Pd/ZSM-5	100	71	4	5		8	3	9

<sup>a</sup> Reaction conditions: 0.2mol BDO, 0.1mol NH<sub>3</sub> (25wt% aqueous solution), 1.8g catalyst, 300 °C, 4Mpa H<sub>2</sub>, 4h.

<sup>b</sup> The catalysts were labeled as xCu-yNi-zPd/ZSM-5 (x, y, z denoted the wt.% of Cu, Ni, Pd to support, respectively.

<sup>c</sup> 1-BePs including: 1-(3-buten-1-yl)-pyrrolidine (1-3-BeP) and 1-(2-buten-1-yl)-pyrrolidine (1-2-BeP)

<sup>d</sup> Other by-products including pyrrolidine, 1-propanol, etc.

<sup>e</sup> Ratio Si/Al of ZSM-5 for Entry 11 and 12 was 25 and 300, respectively.

In order to optimize the reaction conditions for 1-BP, the effects of different parameters were studied in detail using 3Cu-3Ni-0.2Pd/ZSM-5 as catalyst, including the ratio of BDO to NH<sub>3</sub>, reaction atmosphere, pressure and temperature, etc. As shown in Table 2, the selectivity of 1-BP increased with the increase the ratio of BDO to NH<sub>3</sub> (Entry1-3, Table 2), and the highest selectivity could be up to 76% as the BDO: NH<sub>3</sub> was 2:1; whereas, the 1-BP content in the products decreased obviously when the ratio of BDO to NH<sub>3</sub> was further added to 3:1 (Entry 4, Table 2). The reason was that excessive BDO converted to THF smoothly based on the obtained results. Next, the influence of reaction atmosphere on performance was explored. Using N<sub>2</sub> as the reaction atmosphere, only 56% selectivity of 1-BP was obtained (Entry 5, Table 2), while as the reaction atmosphere was changed to  $H_2$ , the 1-BP selectivity could be improved significantly (Entry 6, Table 2). It was found that H<sub>2</sub> played a vital role in the reaction, which was expected to not only maintain the catalyst activity <sup>26, 27</sup>, but also take part in the hydrogenation of 1-BePs. Also, the

synthesis of 1-BP under different hydrogen pressure was carried out. When the pressure of H<sub>2</sub> varied from 1 MPa to 4 MPa (Entry 3, 7-8, Table 2), the selectivity of 1-BP was elevated from 56% to 76% with the complete conversion of BDO; but the selectivity slightly declined to 65% as the pressure of H<sub>2</sub> increased to 6 Mpa (Entry 9, Table 2), because the by-products from the hydrodealkylation and hydrogenation increased under higher hydrogen pressure. Additionally, the reaction temperature was also optimized for this process (Entry 2, 10-12, Table 2). When the reaction temperature was 230 °C, only 35% conversion of BDO with 45% selectivity of 1-BP was gained. Clearly, both the conversion of BDO and selectivity of 1-BP increased with the temperature rising from 250 to 300 °C. The selectivity of 1-BP reached 76% at 300 °C. As the temperature continued to be elevated to 330 °C, the content of 1-BP decreased to 61% with more 1-butanol byproduct. Thus, 1-BP could be yielded via a simple and efficient route from BDO and NH3 over 3Cu-3Ni-0.2Pd/ZSM-5 catalyst at 300 °C for 4 h under a 4 MPa pressure of H<sub>2</sub>. The reuse

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performance of the 3Cu-3Ni-0.2Pd catalyst was tested in second run (Entry 13, Table 2), the selectivity of 1-BP dropped dramatically, which might be due to the catalyst structure was destroyed during the reaction process. But the result was still better than that of the previous report (38% yield).<sup>17</sup>

Table 2 Optimization of the reaction conditions for 1-BP

Entry	T/°C	BDO: NH <sub>3</sub>	Reaction atmosphere	Conv. /%	Sel. /%					
1	300	1: 2	4 Mpa H <sub>2</sub>	35	45					
2	300	1: 1	4 Mpa H <sub>2</sub>	100	15					
3	300	2: 1	4 Mpa H <sub>2</sub>	100	26					
4	300	3: 1	4 Mpa H <sub>2</sub>	100	38					
5	300	2: 1	4 Mpa N <sub>2</sub>	100	56					
6	300	2: 1	4 Mpa H <sub>2</sub>	100	76					
7	300	2: 1	1 Mpa H <sub>2</sub>	100	59					
8	300	2: 1	2 Mpa H <sub>2</sub>	100	69					
9	300	2: 1	6 Mpa H <sub>2</sub>	100	65					
10	330	2: 1	4 Mpa H <sub>2</sub>	100	61					
11	270	2: 1	4 Mpa H <sub>2</sub>	81	58					
12	250	2: 1	4 Mpa H <sub>2</sub>	50	52					
13*	300	2: 1	4 Mpa H <sub>2</sub>	100	63					

Reaction conditions: 1.8g catalyst, 0.2mol BDO, 4h, \* 2<sup>nd</sup> run.

To understand the relationship between catalytic performance and the catalyst properties, the prepared catalysts were characterized by several analytical techniques. Firstly, the contents of Cu, Ni and Pd in the catalysts were determined by atomic absorption spectroscopy (AAS, Table S2). It could be seen that the contents of Cu, Ni and Pd were very close to their theoretical values in all samples. For the 3Cu-3Ni-0.2Pd/ZSM-5, the loading of Cu, Ni and Pd was 2.91%, 2.88% and 0.19% respectively. The loss of Cu and Ni could be detected after use (especially for Cu), but the loss of Pd was not obvious. The Cu loss was likely due to its stronger coordination ability with NH<sub>3</sub>. Thus, the loss of metal species may be one of the reasons for the decrease of catalyst activity after use. The X-ray photoelectron spectroscopy (XPS) studies on the catalyst (3Cu-3Ni-0.2Pd/ZSM-5) were performed to examine the chemical state of active species (Cu, Ni and Pd). As shown in Table S3, the presence of Cu<sub>2</sub>O and NiO could be revealed by XPS analyses, the corresponding binding energy of Cu 2p<sub>3/2</sub> and Ni 2p<sub>3/2</sub> was 932.8 and 855.8 eV, respectively. In addition, the peak appearing at 336.2 eV for Pd 3d<sub>5/2</sub> was detected by XPS technique, revealing that PdO formed on the surface of the developed catalyst.



The XRD of fresh and used 3Cu-3Ni-0.2Pd/ZSM-5 catalysts were illustrated in Fig. 1. The diffraction peaks at 8, 8.9, 14.8 and  $23^{\circ}$  could be attributable to ZSM-5 support (JCPDS: 44-

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0002). Besides, the characteristic peak located at  $37^{\circ}_{VeW}$  (JCPDS: 34-1354) and 43° (JCPDS: 47-1049) wast assigned of 0002 and (111) and NiO (200) facets, respectively, and the results were consistent with XPS analyses. Moreover, there was no evident diffraction pattern for Pd species, which might suggest that Pd species were amorphous or highly dispersed in the catalyst. For the used catalyst, it was found that the intensity of peak for ZSM-5 support became weaker, while the intensity of peak for NiO (200) facets strengthened, which might mean that the structure of the zeolites was destroyed partly during the reaction and the NiO particles agglomerated on the surface of the molecular sieve. The change of the catalyst was one of the reasons for deactivation of the catalyst in the reuse process.

High-resolution transmission electron microscopy (HRTEM) gave the information about morphology and distributions of the active species on the surface of catalyst. From the HRTEM image of 3Cu-3Ni-0.2Pd/ZSM-5 (Fig. 2a), the lattice fringes spacing of 0.24 and 0.21 nm matched well with the Cu<sub>2</sub>O (111) and NiO (200) facets, respectively, which was in accordance with the XRD results according to the corresponding PDF cards (JCPDS: 34-1354, JCPDS: 47-1049). And no detectable lattice fringe for Pd species in the HRTEM image. EDX mapping (Fig. 2b) was conducted to identify the distribution of the elements. The visual inspection of map revealed that Cu, Ni and Pd species clearly overlapped, which indicated that the metal species were uniformly distributed in the catalyst.



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Scheme 2 (a) Synthesis of 1-BP from pyrrolidine and BDO, (b) possible route for 1-BP from NH<sub>3</sub> and BDO

It has been proved that Ni and Cu species displayed excellent catalytic activation in the N-alkylation reaction<sup>12</sup>. Meanwhile, Pd species were effective for both hydrogenation and Nalkylation of amines and alcohols <sup>13</sup>, which was also embodied in this work. And ZSM-5 support might provide appropriate acid-based environment and pore structure 7, 14. In this work, the reducing ambient, introduction of Pd and high dispersion of metals might be conducive to the fine catalytic performance. Besides, a reference reaction of pyrrolidine with BDO was tested under the optimized conditions, and a similar yield (74%) of 1-BP was achieved (Scheme 2a). According to the obtained results, a possible reaction process was speculated (Scheme 2b), namely, pyrrolidine formed firstly through twice N-alkylation between BDO and NH<sub>3</sub> in the presence of ternary catalyst; then BDO as an alkylating agent reacted with the resulted pyrrolidine via another N-alkylation and followed by dehydration to generate 1-BePs<sup>21</sup>; finally, the product 1-BP was obtained after a hydrogenation from 1-BePs. The study on catalytic mechanism of the composite catalyst for this route should be our next work.

In conclusion, the catalytic synthesis of 1-BP and its derivatives was realized using BDO and aqueous ammonia as the raw materials, and the product of 1-BP was emphatically studied. BDO acted as a part of the ring skeleton and the alkylating reagent. The Cu, Ni, and Pd modified ZSM-5 catalyst showed the best catalytic performance, and 76% yield of 1-BP with 5% yield of 1-BePs was obtained under optimization conditions. The preparation of the ternary metals modified catalyst could be carried out by simple impregnation method. It was found that the H<sub>2</sub> atmosphere, introduction of Pd and high dispersion of metals were contributed to the fine catalytic performance. This work provides a promising method with simple and economical characteristics for catalytic synthesis of 1-BP and its derivatives.

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## **Conflicts of interest**

There are no conflicts to declare.

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