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Direct synthesis of a jet fuel range dicycloalkane by the aqueous phase hydrodeoxygenation of polycarbonate†

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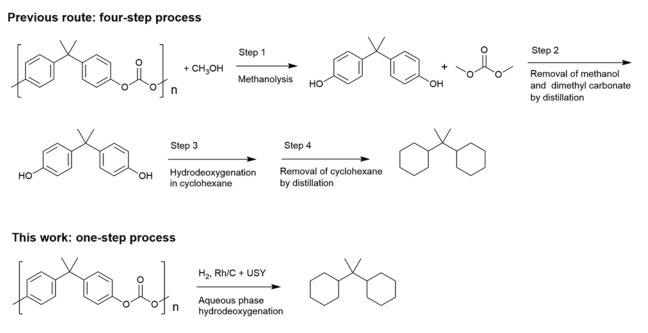
For the first time, propane-2,2-diyldicyclohexane, a jet fuel range C_{15} dicycloalkane was directly produced by the aqueous phase hydrodeoxygenation (APHDO) of polycarbonate (PC). Among the investigated catalyst systems, the mixture of Rh/C and H-USY (denoted as Rh/C + H-USY) exhibited the best performance. Over it, a high yield of propane-2,2-diyldicyclohexane (94.9%) was achieved from the APHDO of pure PC pellets after a reaction was carried out at 473 K and 3.5 MPa H_2 for 12 h. The Rh/C + H-USY catalyst was stable under the investigated conditions. No evident deactivation was observed during three repeated cycles. When we used a chopped DVD disk (a representative of real PC wastes) as the substrate, a high yield (86.9%) of propane-2,2-diyldicyclohexane was obtained under the same reaction conditions. The propane-2,2-diyldicyclohexane as obtained had a high density (0.92 g mL^{-1}) and a high volumetric net heat of combustion (39.6 MJ L^{-1}). As a potential application, it can be blended into jet fuels to improve the range and payload of airplanes.

Introduction

To fulfil the need of environmental protection and sustainable development, the chemical conversion of waste plastics to fuels and chemicals has attracted considerable attention in recent years.[1] Polycarbonate (PC) is one of the most important engineering plastics. Owing to its excellent properties (such as impact resistance, tensile strength, ductility, dimensional stability, and optical clarity), PC has been widely utilized in the production of safety glass, eyeglass lenses, compact discs, construction, containers, *etc.*[2,3] In recent years, numerous efforts have been devoted to the pyrolysis,[4] hydrolysis,[5] alcoholysis,[3,6] aminolysis[7] and glycolysis[8] of PC wastes.

Polycycloalkanes are important components of jet fuel, one of the most demanded liquid transportation fuels in our life. Compared to other jet fuel components, polycycloalkanes have higher density and volumetric heat values. This characteristic can help the airplanes to fly further or carry more load without changing the volume of the fuel tank.[9,10] In the recent study by our group, propane-2,2-diyldicyclohexane, a jet fuel range branched C_{15} dicycloalkane was selectively obtained from the catalytic conversion of polycarbonate (see Scheme 1).[11] However, this process contains at least four steps: (1) the methanolysis of PC to bisphenol A (BPA); (2) the removal of methanol and dimethyl carbonate generated during the methanolysis of PC by distillation; (3) the hydrodeoxygenation of BPA in cyclohexane; (4) the removal of cyclohexane by distillation. To fulfil the need of real applications, an integrated



Scheme 1 Strategy for the aqueous phase hydrodeoxygenation (APHDO) of PC to a jet fuel range high-density C_{15} branched dicycloalkane.

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one-step process is highly expected. Aqueous phase hydrodeoxygenation (APHDO) is a technology, which has been used for the production of renewable fuels from biomass.[12,13] To the best of our knowledge, there is no report on the application of APHDO for the recycling of PC.

In this study, propane-2,2-diyldicyclohexane was first synthesized in a high yield (94.9%) *via* a highly integrated one-step APHDO of PC. Due to its low solubility in water, the propane-2,2-diyldicyclohexane as obtained spontaneously separated from the water after the reaction completed. Compared to our previous route, this new route has numerous advantages, such as fewer steps, lower cost and higher availability of the solvent, lower separation energy consumption and facility investment, *etc.*

Experimental

Materials

In this study, pure poly[2,2-bis(4-hydroxyphenyl)propane carbonate] (PC) pellets (product number: 1609T-11, 2 mm length × 2 mm diameter from Luxi Chemical Group Co., Ltd) were used to simulate the PC waste. The properties of the pure PC pellets are illustrated in Table 1. To check the possibility of a real application, we also used a chopped DVD disk (a representative of real PC) as the substrate for an activity test. Before the activity test, the DVD disk (bought from Mitsubishi Chemical Media Co., Ltd, Chemical composition: 75.8 wt% C; 5.4 wt% H; 18.8 wt% O) was polished with a sandpaper to remove the metal layer, washed thoroughly with ethanol (to remove the organic dye), and cut into 2 mm × 2 mm pieces with scissors.

The Ru/C, Rh/C, Pd/C catalysts and *n*-dodecane (used as an internal standard in this study) were obtained from Aladdin Chemical Reagent Co., Ltd. According to the information from the supplier, the metal contents in the Ru/C, Rh/C and Pd/C catalysts were 5% by weight (denoted as 5 wt%). The metal dispersions of Ru/C, Rh/C, Pd/C catalysts are given in Table S1 of ESI.†

Al_2O_3 was purchased from Sinopharm Chemical Reagent Co., Ltd. $SiO_2–Al_2O_3$ (Al content: 8 wt%) was obtained from Qingdao Hengze silica gel products Co., Ltd. An H-form ultrastable Y-type (H-USY) zeolite (SiO_2/Al_2O_3 molar ratio: 11) was purchased from Nankai University.

For comparison, we also prepared a 5 wt%Rh/H-USY catalyst by the incipient wetness impregnation of H-USY with an aqueous solution of $RhCl_3$. Before being used in the activity tests, the Rh/H-USY catalyst was reduced in a hydrogen flow at 573 K for 2 h, cooled down in the hydrogen flow to room temperature, and then passivated in a 1% O_2/N_2 flow for 0.5 h.

Table 1 The properties of pure PC pellets used in the activity tests

	Pure PC pellet	Chopped DVD disk
Average molecular weight[a]	3.6×10^4	2.6×10^4
Melt point[b] (K)	456–461	428–440
Decomposition temperature[c] (K)	718	703

[a] Measured by a waters 1525 & Agilent PL-GPC220 gel permeation chromatography (GPC). [b] Measured by an X-5 micro melting point apparatus (Beijing Tech Instrument Co. Ltd). [c] Measured by a thermogravimetric-differential scanning calorimetry (TG-DSC).

Characterization

The specific BET surface areas (S_{BET}) and pore sizes of the investigated catalysts were measured by N_2-physisorption at 77 K using a Micromeritics ASAP 2010 apparatus.

The acid amounts and acid strengths of the solid acids were characterized by NH_3-chemisorption and NH_3-TPD, respectively. Before each test, 0.1 g sample was placed in a quartz reactor, pre-treated in a He flow at 773 K for 0.5 h, and cooled down in He flow to 373 K. After the stabilization of the base line, the pulses of NH_3 (1 mL) were dosed in the reactor until saturation. The amount of acid sites on the catalyst was calculated by the adsorption of NH_3 during the test. Subsequently, the desorption of NH_3 was conducted in He flow from 373 K to 1173 K at a heating rate of 10 K min^{-1}. The desorbed NH_3 molecules were detected by a mass spectrometry (MS) OmniStar equipped with the software QuadStar.

Activity tests

The APHDO of PC was carried out in a 100 mL stainless steel batch reactor (Parr 4848) under hydrogen atmosphere. For each test, 1 g PC, 40 mL water, 0.1 g M/C (M = Pd, Ru, Rh) and/or 0.1 g H-USY were used. After purging the reactor with argon for three times, the reactor was charged with hydrogen to 3.5 MPa, the mixture was stirred at 473 K for 12 h, and then quenched to room temperature with cool water. The products generated during the APHDO of PC were solved in tetrahydrofuran, and quantitatively analyzed by an Agilent 7890A GC. The PC conversions and the yields of C_{15} oxygenates and propane-2,2-diyldicyclohexane were calculated according to following equations:

$$\text{Conversion of PC}(\%) = (\text{Initial weight of pure PC pellets } (\text{or chopped DVD disk}) - \text{the weight of residual PC pellets } (\text{or residual chopped DVD disk}))/(\text{Initial weight of pure PC pellets } (\text{or chopped DVD disk})) \times 100\%$$

Yield of C_{15} oxygenates or propane-2,2-diyldicyclohexane (%) = (Mole of C_{15} oxygenates or propane-2,2-diyldicyclohexane generated during the activity test)/(Theoretical mole of oxygenates or the propane-2,2-diyldicyclohexane which should be produced from the complete conversion of the PC (or chopped DVD disk)) × 100%.

Results and discussion

First, we studied the APHDO of pure PC pellets (denoted as PC) over the commercial Rh/C and H-USY catalysts, and their physical mixture (denoted as Rh/C + H-USY). It can be seen from Fig. 1 that the Rh/C catalyst was effective for the degradation of PC. Moreover, PC was completely converted, and

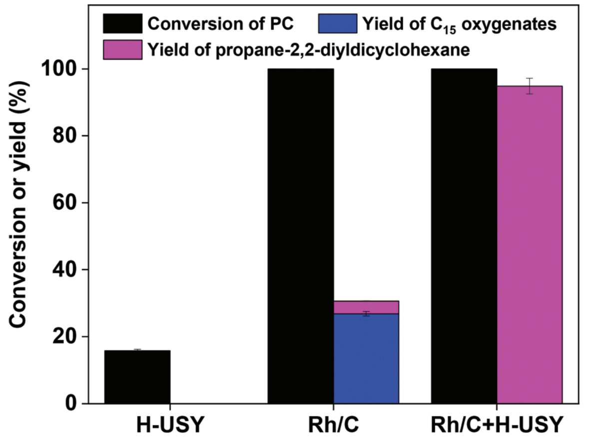
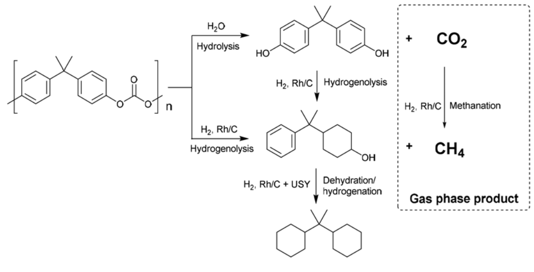


Fig. 1 Conversions of PC and the yields of different products over the Rh/C, H-USY and Rh/C + H-USY catalysts. Reaction conditions: 473 K, 3.5 MPa H_2, 12 h; 1 g pure PC pellets, 0.1 g Rh/C and/or 0.1 g H-USY catalyst and 40 mL H_2O were used in each test.

26.9% yield of C_{15} oxygenates and 3.8% yield of propane-2,2-diyldicyclohexane were achieved after the reaction carried out at 473 K for 12 h. According to the GC-MS analysis illustrated in Fig. S1–S4 of ESI,† the C_{15} oxygenates as obtained were mainly composed of bisphenol A (BPA) and 4-benzylcyclohexan-1-ol. From the reaction pathways we proposed in Scheme 2, BPA was generated from the hydrolysis of PC, while 4-benzylcyclohexan-1-ol was generated from the partial hydrodeoxygenation of BPA. The H-USY zeolite also worked for the degradation of PC. However, its activity was much lower than that of Rh/C. For the H-USY catalyst, a low PC conversion (15.8%) was achieved, and no C_1–C_{15} oxygenate (or hydrocarbon) was obtained under the same reaction conditions as we used for the Rh/C catalyst. From the analysis of the gas phase products (see Fig. S5 in ESI†), both CO_2 and CH_4 were generated from the degradation of PC over the Rh/C catalyst. In contrast, CO_2 was identified as the only gaseous product when H-USY was used as the catalyst. It can be seen from Scheme 2 that CO_2 was produced from the hydrolysis of PC, while CH_4 was generated by the hydrogenolysis of the carbonate groups in PC (or the methanation of CO_2). Based on the above information, it can be seen that the presence of the Rh/C catalyst promoted the hydrogenation of carbonate (or CO_2) and BPA, which may be the reason for the relatively higher PC conversion over the Rh/C catalyst. It was very interesting that the main product switched to propane-2,2-diyldicyclohexane when we used Rh/C + H-USY as the catalyst. Over it, a high yield (94.9%) of propane-2,2-diyldicyclohexane was obtained from the APHDO of PC under the investigated conditions. To the best of our knowledge, this is the first report about the direct production of propane-2,2-diyldicyclohexane by the APHDO of PC.

It is worth mentioning that propane-2,2-diyldicyclohexane as obtained had a low solubility in water. As the result, it is spontaneously separated from water after the reaction was finished (see Fig. S6 in ESI†). In real applications, this characteristic may save the energy consumption for the removal of the solvent. Based on the information illustrated in Table 2, the density and volumetric net heat of combustion (NHOC) of propane-2,2-diyldicyclohexane are evidently higher than those of the currently used jet fuels and rocket fuels (for example JP-8 fuel and RP-1 fuel). As a potential application, it can be blended into jet fuels or rocket fuels to increase the range and payload of airplanes or rockets. Moreover, it is noticed that the density and volumetric NHOC of propane-2,2-diyldicyclohexane were also comparable to those of JP-10 fuel and RJ-4 fuels, two classic single-component advanced jet fuels, that are widely used for military jets, missiles, and supersonic combustion ramjets. It is known that the RJ-4 and JP-10 fuels are synthesized with methylcyclopentadiene and cyclopentadiene, which are obtained from fossil fuels in very low yields.[17,18] As a result, the JP-10 fuel has higher price than that of pure PC (~7000 USD ton^{-1} *vs.* ~2000 USD ton^{-1}).[14] The price of RJ-4 is even higher.[18] Therefore, the production of propane-2,2-diyldicyclohexane from the APHDO of PC should be economically viable if it can partially substitute the currently used high-density aviation fuels.

To figure out the reason for the excellent performance of the Rh/C + H-USY catalyst, and the respective roles of Rh/C and H-USY in the APHDO of PC to propane-2,2-diyldicyclohexane, we studied the catalytic performances of H-USY, Rh/C and Rh/C + H-USY for the APHDO of diphenyl carbonate (a good model compound that has a similar chemical structure to that of PC) and BPA (*i.e.*, the hydrolysis product of PC). The results are shown in Fig. 2. In the absence of any catalyst, diphenyl carbonate can be converted to phenol by hydrolysis. This



Scheme 2 Reaction pathways for the generation of different products from the APHDO of PC.

Table 2 Properties of propane-2,2-diyldicyclohexane and the currently used aviation fuels

	Density ($g\ mL^{-1}$)	Volumetric NHOC[a] ($MJ\ L^{-1}$)
JP-8 fuel[14]	0.793	34.5
RP-1 fuel[15]	0.81	35.6
JP-10 fuel[10]	0.936	39.4
RJ-4 fuel[10]	0.927	36.2
Propane-2,2-diyldicyclohexane[16]	0.916	39.6

[a] Volumetric NHOC: volumetric net heat of combustion.

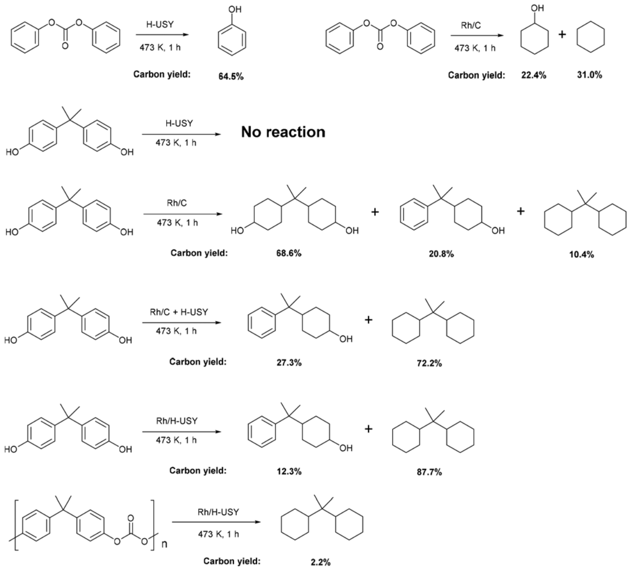


Fig. 2 Yields of different products from the APHDO or hydrodeoxygenation (HDO) of diphenyl carbonate (or BPA) over the Rh/C, H-USY and Rh/C + H-USY catalysts. Reaction conditions: 473 K, 3.5 MPa H_2; 4 mmol model compound, 0.1 g Rh/C and/or 0.1 g H-USY catalyst and 40 mL H_2O (or hexane) were used in each test.

could be comprehended because high-temperature water can act as a catalyst for some acid-catalyzed reactions.[21] Analogously, phenol was obtained as the main product from the APHDO of diphenyl carbonate over H-USY after the reaction was carried out at 473 K for 1 h. Over H-USY, the phenol yield (64.5%) was evidently higher than the one achieved in the absence of a catalyst under the same reaction conditions (16.2%). This result indicates that H-USY had some kind of promotion effects on the hydrolysis of carbonate. However, no cyclohexane or benzene was detected in the product over H-USY. The low activity of H-USY for the APHDO of the phenolic compound was further confirmed when we used BPA as the substrate. In contrast, cyclohexanol and cyclohexane were identified as the major products from the APHDO of diphenyl carbonate over the Rh/C catalyst. Analogously, 4,4′-(propane-2,2-diyl)dicyclohexanol, 4-(2-phenylpropan-2-yl)cyclohexanol and propane-2,2-diyldicyclohexane were obtained from the APHDO of BPA over the Rh/C catalyst. Based on these results, we could see that H-USY could only promote the hydrolysis of carbonate. However, the presence of a metal catalyst was indispensable for the further hydrodeoxygenation of phenol or BPA. This can be comprehended because the Ar–OH bond has a bond dissociation enthalpy of 465 kJ mol^{-1}, which is even higher than that of the C–H bond in methane (439 kJ mol^{-1}).[19] As a result, the hydrogenation of the benzene ring is favorable for hydrodeoxygenation. Compared to the Rh/C catalyst, the Rh/C + H-USY catalyst had a significantly higher activity for the APHDO of BPA (see Fig. 2). Over it, a much higher yield of propane-2,2-diyldicyclohexane (72.2% *vs.* 10.4%) was achieved under the same reaction conditions. Therefore, we believe that there was a synergistic effect between the Rh/C and H-USY catalyst in the APHDO of BPA to propane-2,2-diyldicyclohexane. This phenomenon can be rationalized because acid sites are important for the dehydration of cyclic diols or alcohols from the hydrogenation (or partial hydrodeoxygenation) of BPA. According to a previous study by Huber *et al.* on the reaction mechanism for APHDO,[20] the acid-catalyzed dehydration of an alcohol (or polyol) followed by hydrogenation over metal sites is the major pathway for the APHDO of oxygenates. It is worth mentioning that the solvent also played an important role in the APHDO of BPA over the Rh/C catalyst. When we used hexane as the solvent, evidently lower yields of hydrodeoxygenation (HDO) products (*i.e.* 4-(2-phenylpropan-2-yl) cyclohexanol and propane-2,2-diyldicyclohexane) were achieved over the Rh/C catalyst than those obtained when we used water as the solvent. Such a solvent effect was not apparent when we used H-USY + Rh/C as the catalyst. Based on these results, we can comprehend the APHDO activity of the Rh/C catalyst by two reasons: (1) the beneficial effect of high-temperature water. As what has been suggested in some studies,[21] high-temperature water can work as an acid catalyst to promote dehydration, which may be one reason for the APHDO activity of the Rh/C catalyst; (2) and the other is the acidity of the Rh/C catalyst. Based on the NH_3-TPD result shown in Fig. S7 of ESI,† the Rh/C catalyst had some acidity. This may be the reason for the generation of HDO products over the Rh/C catalyst even when the reaction was carried out in hexane. According to the literature,[13,22] the acidity of the Rh/C catalyst can be attributed to the Rh oxide (Lewis acidic sites) generated by the contact of the catalyst with air.

It is worth mentioning that the Rh/H-USY catalyst was more active than the Rh/C + H-USY catalyst for the APHDO of BPA. However, this catalyst was inactive for the APHDO of PC. After the reaction was carried out under the investigated conditions for 12 h, a low PC conversion (2.7%) was achieved over the Rh/H-USY catalyst, and no propane-2,2-diyldicyclohexane was identified in the product. This result could be rationalized because the loading of Rh may block some pores on the surface of H-USY, and restrain the hydrolysis of PC. To verify this speculation, we compared the activity of Rh/H-USY and H-USY for the degradation of PC under an argon atmosphere. As we expected, a much lower PC conversion (2.8%) was obtained over Rh/H-USY than that over H-USY (13.6%). Furthermore, we also studied the activity of Rh/C and Rh/C + H-USY catalysts for the degradation of PC under argon atmosphere. Under the same conditions, the PC was completely converted. However, no C_{15} oxygenate was identified in the products. Based on these results, it can be seen that the presence of the Rh/C catalyst is favorable for the degradation of PC. This result may be rationalized by the acidity of the Rh/C catalyst.

For comparison, we studied the catalytic performances of other M/C + H-USY (M = Pd or Ru) catalysts for the APHDO of PC under the same reaction conditions as we used for Rh/C + H-USY (see Fig. 3 and Fig. S8 in ESI†). As we expected, the Ru/C + H-USY and Pd/C + H-USY catalysts were also active for the APHDO of PC to propane-2,2-diyldicyclohexane. However, their

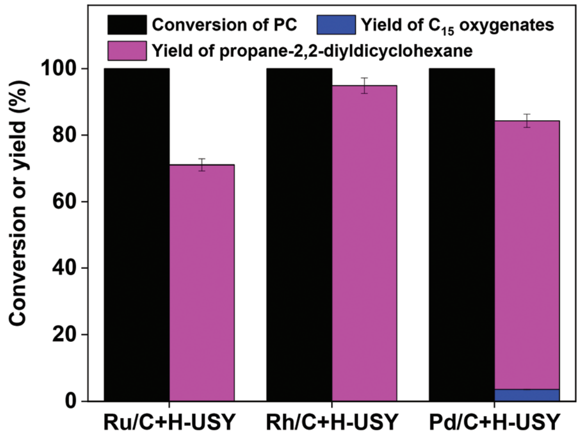


Fig. 3 Conversions of PC and the yields of different products over the M/C (M = Ru, Rh or Pd) + H-USY catalysts. Reaction conditions: 473 K, 3.5 MPa H_2, 12 h; 1 g pure PC pellets, 0.1 g M/C, 0.1 g H-USY and 40 mL H_2O were used in each test.

activities were evidently lower than that of Rh/C + H-USY. According to the literature,[23] the higher activity of Rh/C + H-USY can be explained by the higher activity of Rh-based catalysts for the activation of hydrogen and the hydrogenation of carboxyl groups (or esters).

The effect of solid acids was explored as well. From Fig. 4, it can be seen that the Rh/C + H-USY catalyst was more active than the Rh/C + SiO_2–Al_2O_3 and Rh/C + Al_2O_3 catalysts for the APHDO of PC. Based on the characterization results illustrated in Fig. 5 and Table 3, it can be seen that the H-USY zeolite had a higher acid strength, bigger surface area and amount of acid sites than those of SiO_2–Al_2O_3 and Al_2O_3. This may be the reason for the excellent performance of the Rh/C + H-USY catalyst.

The effects of the reaction temperature and the reaction time on the catalytic performance of the Rh/C + H-USY were also investigated. From the results illustrated in Fig. 6 and 7, the highest propane-2,2-diyldicyclohexane yield was achieved over the Rh/C + H-USY catalyst after the reaction was carried out at 473 K for 12 h.

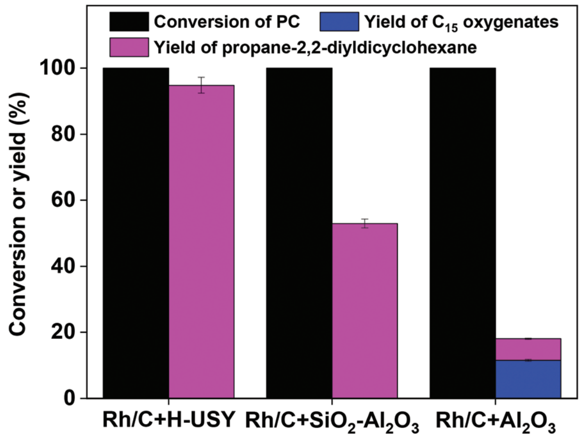


Fig. 4 Conversions of PC and the yields of different products over the Rh/C + H-USY, Rh/C + SiO_2–Al_2O_3 or Rh/C + Al_2O_3 catalysts. Reaction conditions: 473 K, 3.5 MPa H_2, 12 h; 1 g pure PC pellets, 0.1 g Rh/C, 0.1 g solid acid and 40 mL H_2O were used in each test.

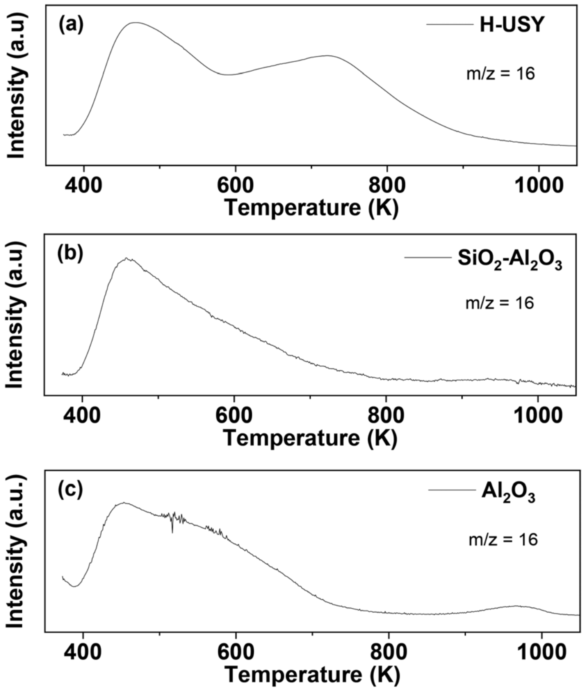


Fig. 5 NH_3-TPD profiles of the H-USY (a), SiO_2–Al_2O_3 (b) and Al_2O_3 (c).

To fulfil the need of real applications, we also checked the reusability of the Rh/C + H-USY catalyst. To achieve this, the Rh/C + H-USY catalyst was repeatedly used under the reaction conditions that have been optimized for the pure PC pellets. After each usage, the catalyst was separated from the reaction system by filtration, thoroughly washed by ethanol, and dried in vacuum at 333 K for 8 h. From the result illustrated in Fig. 8, the Rh/C + H-USY catalyst was stable under the investigated conditions. No evident decrease in the activity was observed after it was repeatedly used for three times. The slight decrease in propane-2,2-diyldicyclohexane at the third usage can be rationalized by the loss of the catalyst during the recycling of the catalyst by filtration. Based on this result, we believe that the coke that may be formed on the catalyst (by the unreacted molecules) had no evident effect on the catalytic performance of the Rh/C + H-USY catalyst.

Considering the great prospect of this method for the production of a jet fuel range high-density dicycloalkane from PC

Table 3 Specific surface areas (S_{BET}), the acid amounts and pore sizes of the solid acids used in this study

Catalyst	S_{BET} [a] (m^2 g^{-1})	Acid amount[b] (mmol g^{-1})	Pore size[a] (nm)
H-USY	657	0.880	0.74 × 0.74 (ref. 24)
SiO_2–Al_2O_3	325	0.322	4.5
Al_2O_3	204	0.430	8.9

[a] Measured by N_2-physisorption. [b] Measured by NH_3-chemisorption.

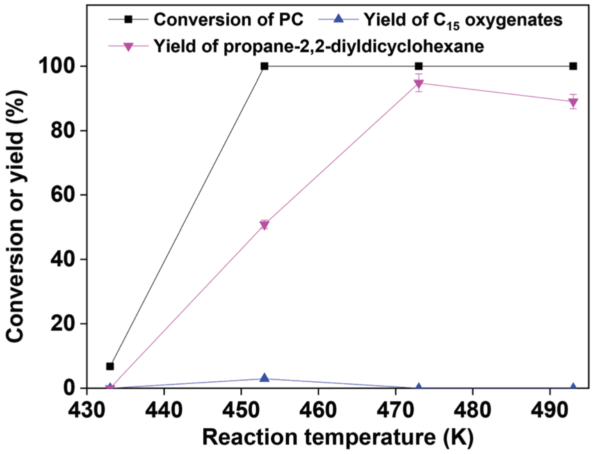


Fig. 6 Conversions of PC and the yields of propane-2,2-diyldicyclohexane over the Rh/C + H-USY catalyst as the function of the reaction temperature. Reaction conditions: 3.5 MPa H_2, 12 h; 1 g pure PC pellets, 0.1 g Rh/C, 0.1 g H-USY and 40 mL H_2O were used in each test.

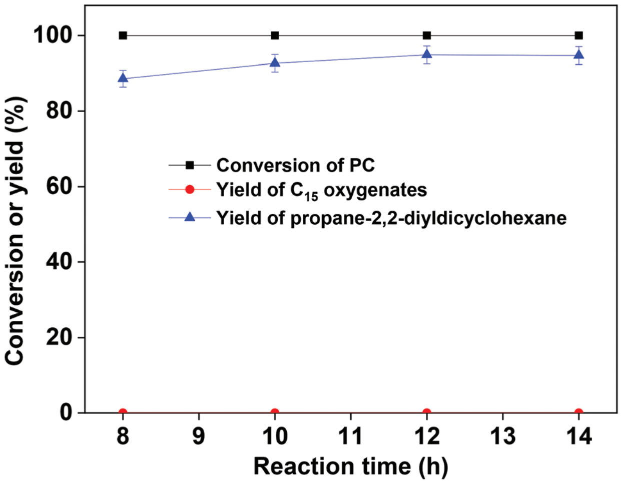


Fig. 7 Conversions of PC and the yields of propane-2,2-diyldicyclohexane over the Rh/C + H-USY catalyst as the function of the reaction time. Reaction conditions: 473 K, 3.5 MPa H_2; 1 g pure PC pellets, 0.1 g Rh/C, 0.1 g H-USY and 40 mL H_2O were used in each test.

waste, we also studied the catalytic performance of the Rh/C + H-USY catalyst for the APHDO of the chopped DVD disk, which can be considered as a representative of real PC wastes. The calculation methods for the conversion of the chopped DVD disk and the yield of different products were the same as what have used for the pure PC pellets, supposing the purity of the chopped DVD disk was 100%. Under the same reaction conditions for the pure PC pellets, a high yield (86.9%) of propane-2,2-diyldicyclohexane was achieved. For comparison, we also did a blank experiment in argon atmosphere without using any catalyst. Under the same reaction conditions as we used for the Rh/C + H-USY catalyst, 16.8% conversion of the chopped DVD disk was achieved. However, no C_{15} oxygenates or hydrocarbons were identified in the product.

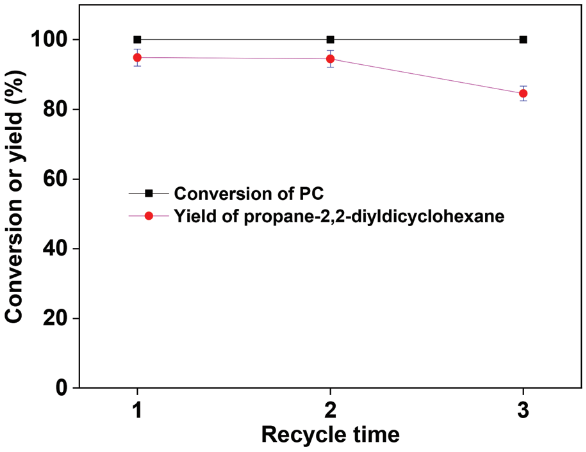


Fig. 8 Conversions of PC and the yields of propane-2,2-diyldicyclohexane over the Rh/C + H-USY catalyst as the function of the recycle time. Reaction conditions: 473 K, 3.5 MPa H_2, 12 h; 1 g pure PC pellets, 0.1 g Rh/C, 0.1 g H-USY and 40 mL H_2O were used in each test.

Conclusions

In summary, APHDO was proved to be a promising method for the direct production of a jet fuel range high-density dicycloalkane from PC. Rh/C + H-USY was found to be an active and stable catalyst for this reaction. Under the investigated conditions, high yields (94.9% and 86.9%) of propane-2,2-diyldicyclohexane were achieved from the APHDO of pure PC pellets and the chopped DVD disk. In the APHDO of PC, the Rh/C catalyst promoted the hydrogenolysis and hydrogenation reactions, while the H-USY and high-temperature water promoted the hydrolysis of PC and the dehydration of alcohols (or diols) from the hydrodeoxygenation (or hydrolysis/hydrogenation) of PC. All of these effects led to the high activity of the Rh/C + H-USY catalyst in the APHDO of PC. The propane-2,2-diyldicyclohexane as obtained had a high density and a good volumetric NHOC. As a potential application, it can be blended into jet fuels to increase the range and the payload of airplanes or rockets. This study opens a new general strategy for the direct conversion of PC wastes to high-quality jet fuels. Further studies regarding the utilization of non-noble metal catalysts in the APHDO of PC wastes to jet fuel range dicycloalkanes are ongoing in our laboratory.

Conflicts of interest

There are no conflicts to declare.

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