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## **Electronic Supplementary Information**

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## Theoretical specific rate was estimated as follows:

It is assumed that all the Ni particles on the three samples are spherical and with a uniform size distribution. Then, the specific rate on per mole Ni (abbreviated as R, with a unit of mol<sub>phenol</sub> mol<sub>Ni</sub><sup>-1</sup> h<sup>-1</sup>) would be:

$$R = V_m/V_1 * S_1 * R = V_m/(4\pi r^3/3) * 4\pi r^2 * R = 3V_mR/r$$

V<sub>m</sub>: the molar volume of Ni;

V<sub>1</sub>: the volume of one Ni particle;

S<sub>1</sub>: the surface area of one Ni particle;

R: the specific rate on per unit Ni surface area, with a unit of mol<sub>phenol</sub> m<sup>-2</sup> h<sup>-1</sup>;

r: the mean radius of the Ni particles;

Therefor, for the sample Ni/OND-R with a mean size of Ni of 8.9 nm, the R should be:

$$R = 3V_mR/(8.9/2) = 59 \text{ mol}_{phenol} \text{ mol}_{Ni}^{-1} \text{ h}^{-1}$$
, scilicet  $3V_mR = 59*(8.9/2) = 262.55$ .

By assuming the *R* of the Ni/ND-R and Ni/NND-R is the same as that of the Ni/OND-R, then, the calculated R for them should be:

For the Ni/ND-R with a mean Ni particle size of Ni of 12.5 nm: the calculated  $R = 3V_mR/r = 3V_mR/(12.5/2) = 262.55/(12.5/2) = 42 \text{ mol}_{phenol} \text{ mol}_{Ni}^{-1} \text{ h}^{-1}$ ;

For the Ni/NND-R with a mean Ni particle size of Ni of 16 nm: the calculated  $R = 3V_mR/r = 3V_mR/(16/2) = 262.55/(16/2) = 33 \text{ mol}_{phenol} \text{ mol}_{Ni}^{-1} \text{ h}^{-1}$ ;

Table S1 XRD patterns fitting results of supports and Ni/support-R samples.

Samples	Ι	Diamond(111	)	Ni(111)		
	Position (°)	FWHM <sup>[a]</sup> (°)	Size <sup>[b]</sup> (nm)	Position (°)	FWHM <sup>[a]</sup> (°)	Size <sup>[b]</sup> (nm)
NND	43.69	2.34	3.6			
Ni/NND-R	43.69	2.34	3.6	44.35	0.53	16
OND	43.69	2.34	3.6			
Ni/OND-R	43.69	2.34	3.6	44.35	0.95	8.9
ND	43.69	2.34	3.6			
Ni/ND-R	43.69	2.34	3.6	44.19	0.68	12.5

<sup>[</sup>a] The full width at half maximum. [b] Particle size estimated according to the Scherrer formula.

As shown in Figure 2a, the XRD patterns of fresh supports NND ND and OND are basically the same. The diamond(111) diffraction peak parameters of each patterns were further calculated by the Scherrer formula, and the results showed that the particle size of each fresh support was the same, which was 3.6 nm (Table S1). The recovered supports were obtained by acid dissolving the nickel species of the Ni/support-R samples, and the collected supports were denoted as recovered supports. For example, recovered-OND was obtained by acid treating of the Ni/OND-R sample. It can be seen from the Figure 2a that the patterns of each recovered supports are basically the same as the corresponding fresh support, indicating that the support exhibits excellent anti-sintering properties during the calcination and reduction treatment that the catalyst preparation process involved, consequently, the particle size has not grown. Based on this result, during the fitting process of the XRD patterns of the Ni/support-R samples (Figure 2c), the diamond(111) diffraction peak parameters of all Ni/support-R samples were set to be consistent values: peak located at 43.69°, with a full width at half maximum (FWHM) of 2.34°. Then, the Scherrer formula calculation was performed on the fitted diffraction peaks of the Ni(111) to figure out the size of Ni species (calculation results are shown in Table S1).

Table S2  $H_2$ -TPR results of ND support, Ni/NND, Ni/OND and Ni/ND.

Samples	Reduction of	nickel species	Reduction of support		
	Peak 1 position (°C)	Peak 1 position (°C)	Peak position (°C)	Area <sub>sample</sub> /Area <sub>ND</sub> [a]	
ND			615	1.0	
Ni/NND	345		570	0.5	
Ni/OND		416	570	0.9	
Ni/ND	338	416	570	0.8	

<sup>[</sup>a] The ratio of the support reduction peak area of each Ni/support samples to the reduction peak area of ND

Table S3 XPS-O1s result of Ni/ND-R, Ni/OND-R and Ni/NND-R.

Samples -		Binding Energy (eV)				Area <sub>sample</sub> /Area <sub>Ni/ND-R</sub> [b]
	$O_a$	$O_b$	$O_c$	$O_{d}$	$O_d/O_{all}^{[a]}$	Al Ca <sub>sample</sub> /Al Ca <sub>Ni/ND-R</sub>
Ni/NND-R	532.7	531.4	529.8		0/100	1.0
Ni/OND-R	532.7	531.3	529.8	528.8	9/91	0.7
Ni/ND-R	532.7	531.3	529.8	528.8	5/95	0.9

<sup>[</sup>a] The ratio of the O<sub>d</sub> area to the total oxygen species area. [b] The ratio of the oxygen species area of each Ni/support-R samples to that of the Ni/NND-R.