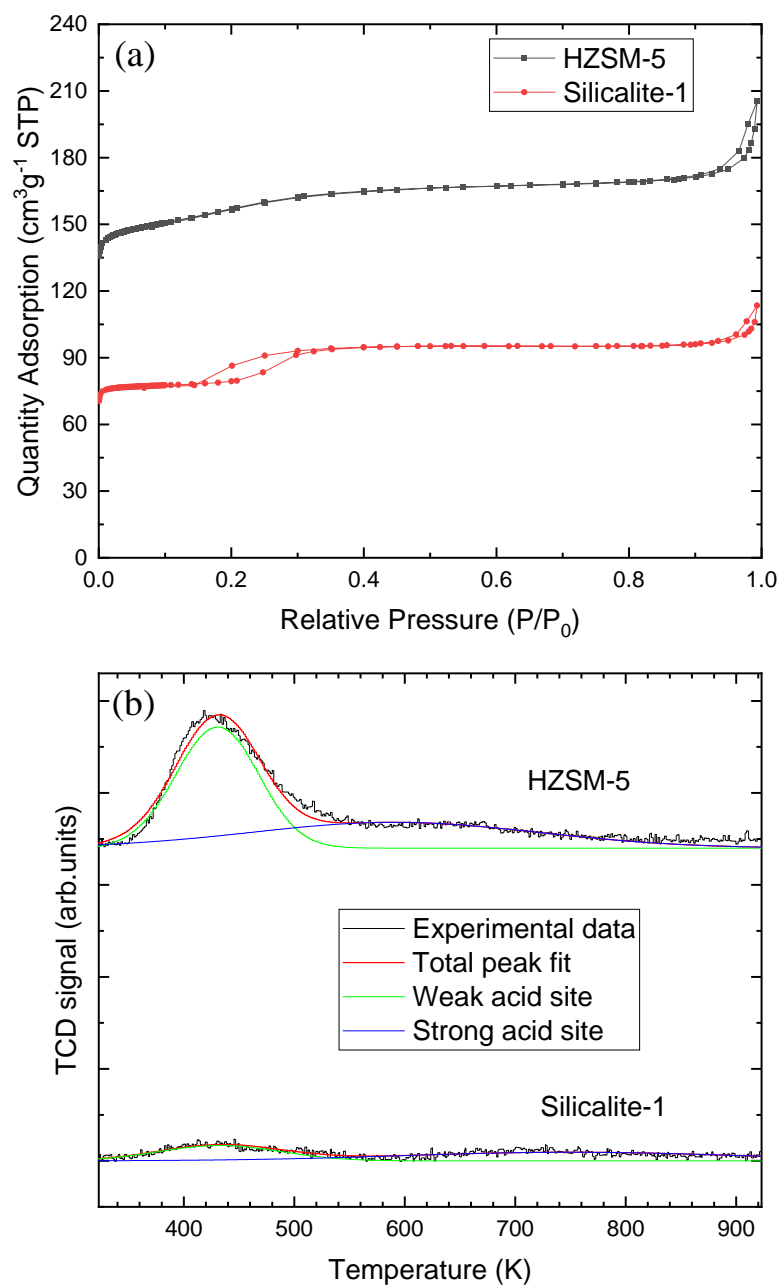
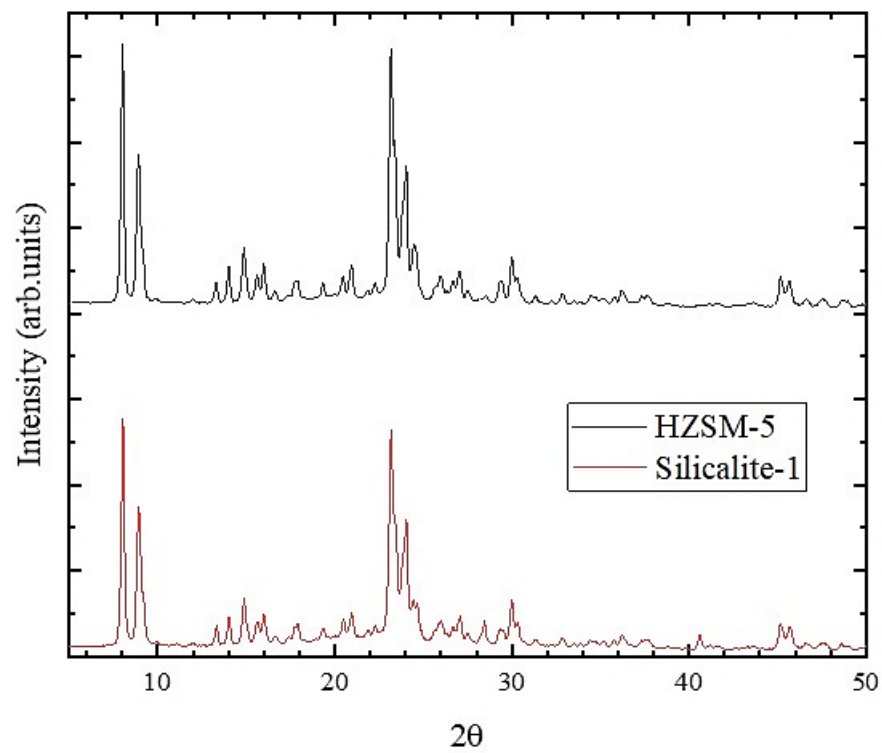


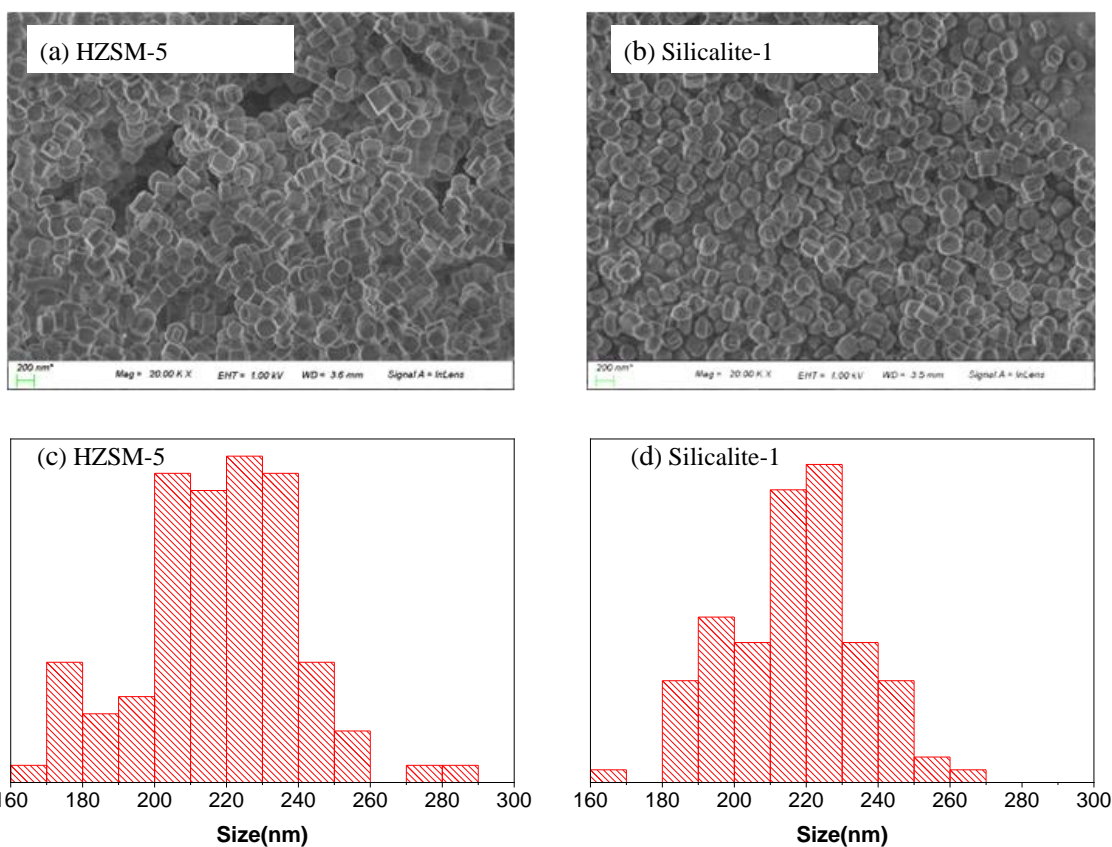
**Figure 1.** Optical images of levitated droplets of JP-10 containing (a) 5 wt % of HZSM-5 zeolites (hereafter: zeolite-JP-10), (b) 5 wt % of HZSM-5 zeolites and 1 wt % Al NPs (hereafter: zeolite-Al-JP-10), (c) 1 wt % of HZSM-5 zeolites and 5 wt % span@80 (hereafter: zeolite-JP-10-surfactant), and (d) 1 wt % of HZSM-5zeolites, 1 wt % Al NPs, and 5 wt % span@80 (hereafter: zeolite-Al-JP-10-surfactant).



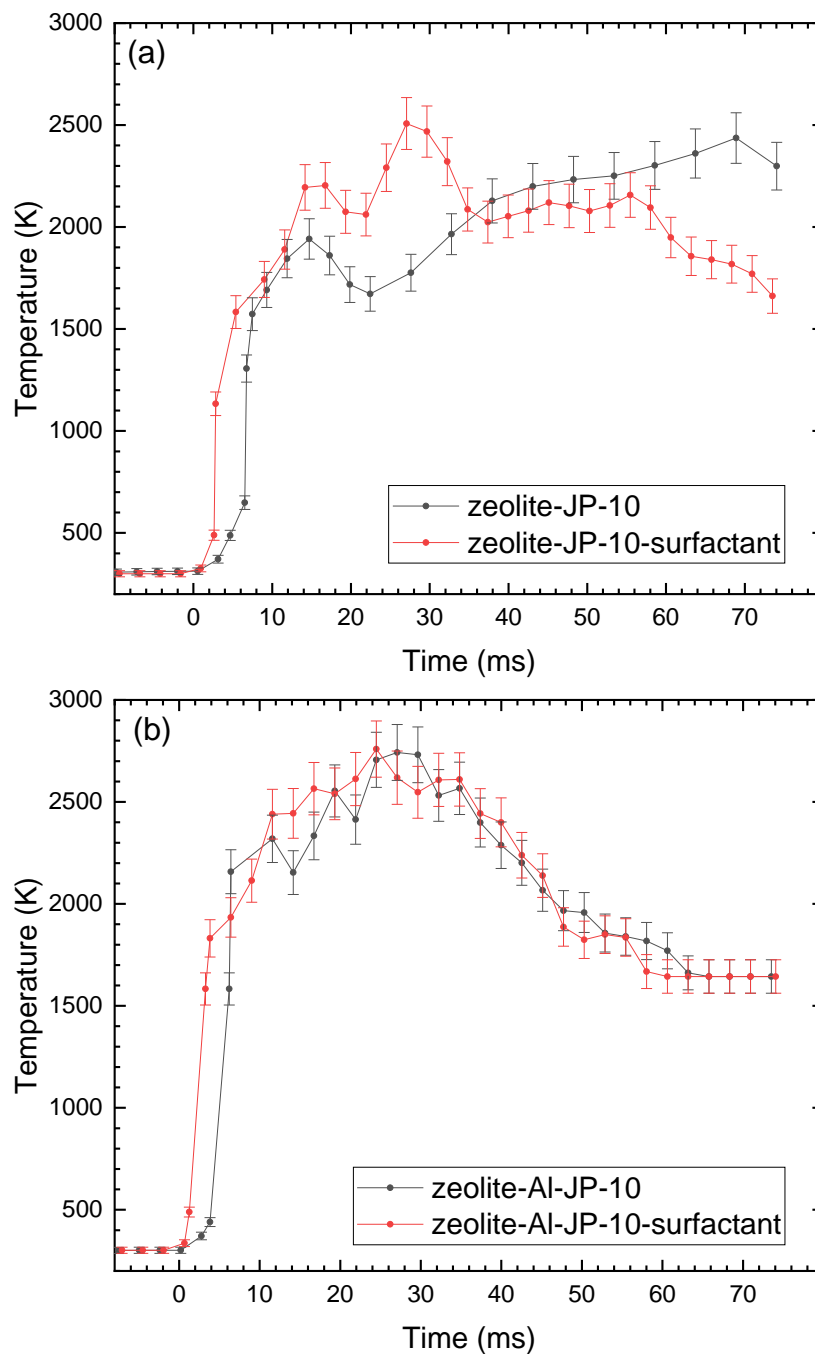
**Figure 2.** (a) Nitrogen ( $N_2$ ) adsorption isotherms and (b) ammonia ( $NH_3$ ) TPD-MS profiles of  $H^+$ -MFI-65 (hereafter: HZSM-5), and MFI- $\infty$  (hereafter: Silicalite-1). The ammonia TPD profiles can be deconvoluted to three peaks: total peak fit (red), weak (green) and strong (blue) acidic sites. The nitrogen adsorption isotherm of HZSM-5 was offset by  $50\text{ cm}^3 g^{-1}$ .



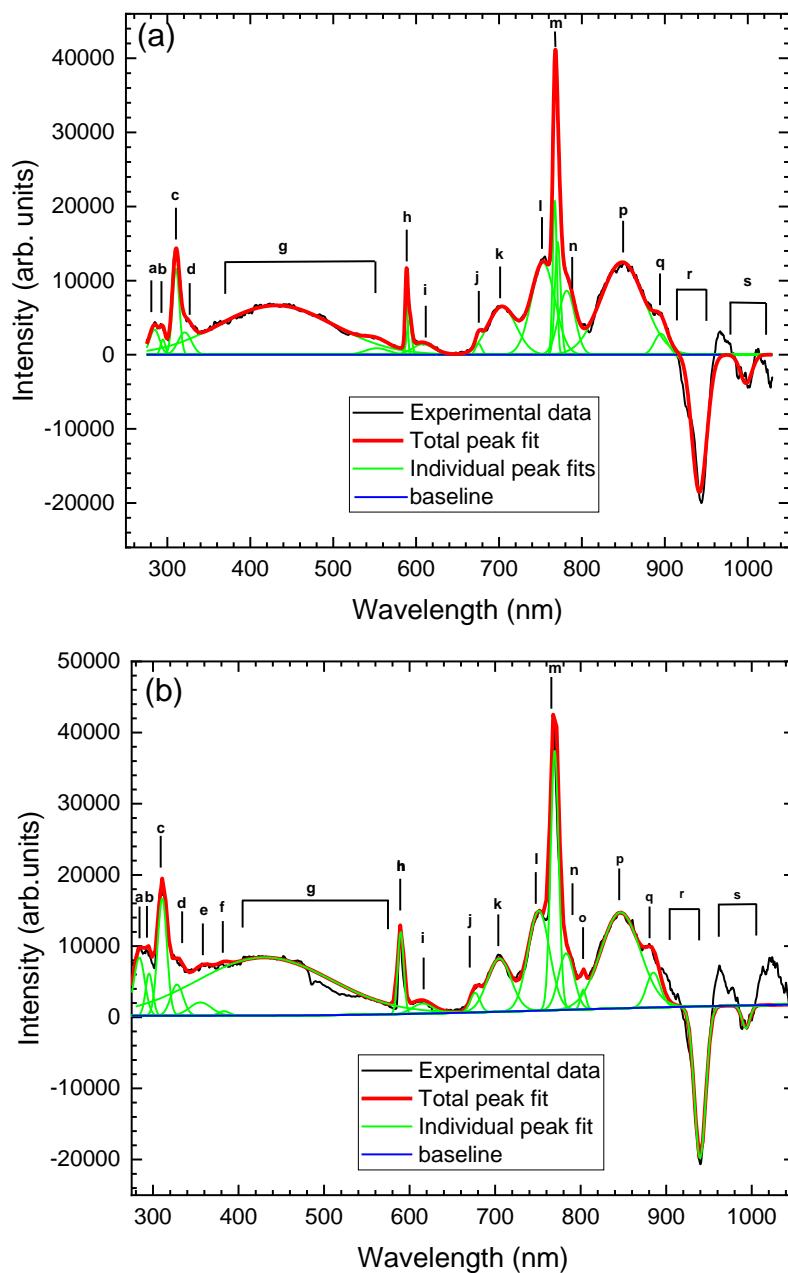
**Figure 3.** X-ray diffraction patterns of HZSM-5 and Silicalite-1.



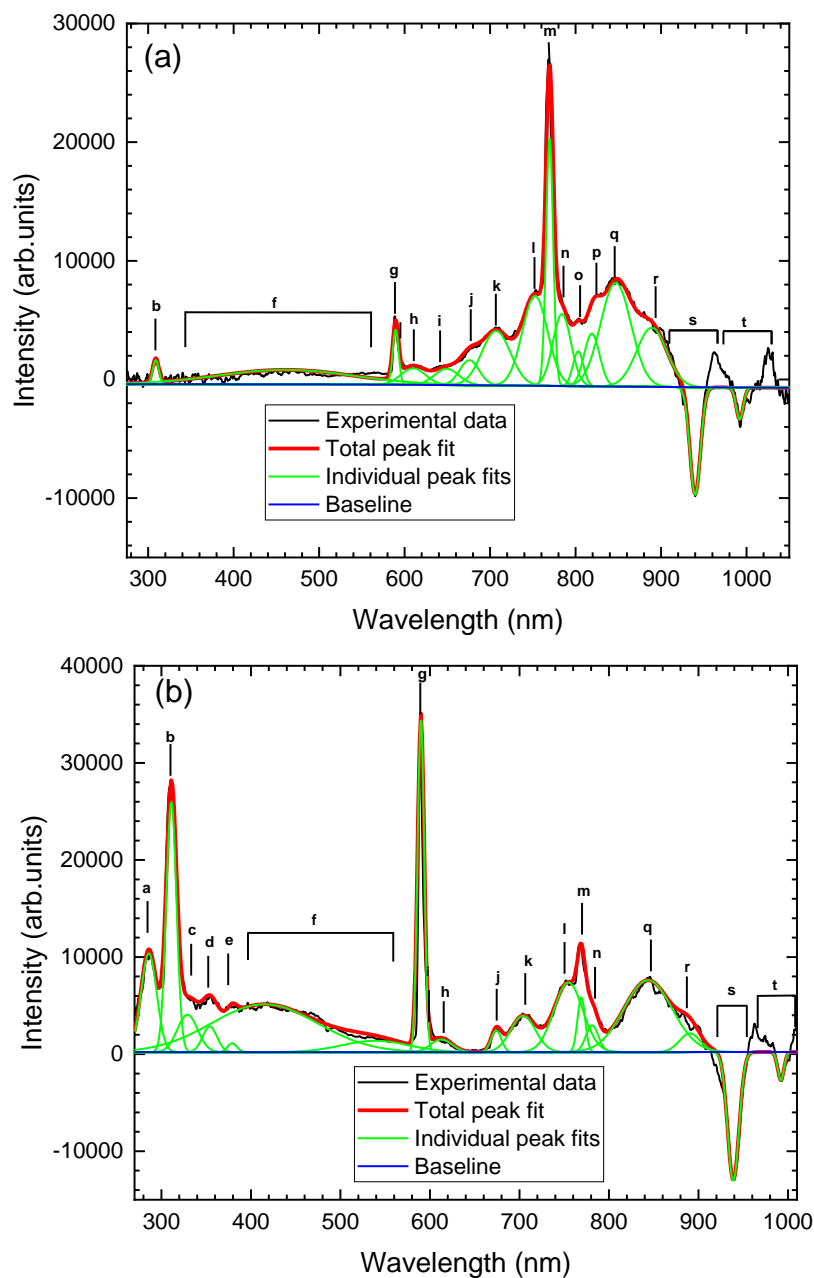
**Figure 4.** Scanning electron microscope images of (a) HZSM-5 and (b) Silicalite-1, and (c-d) their respective histograms.



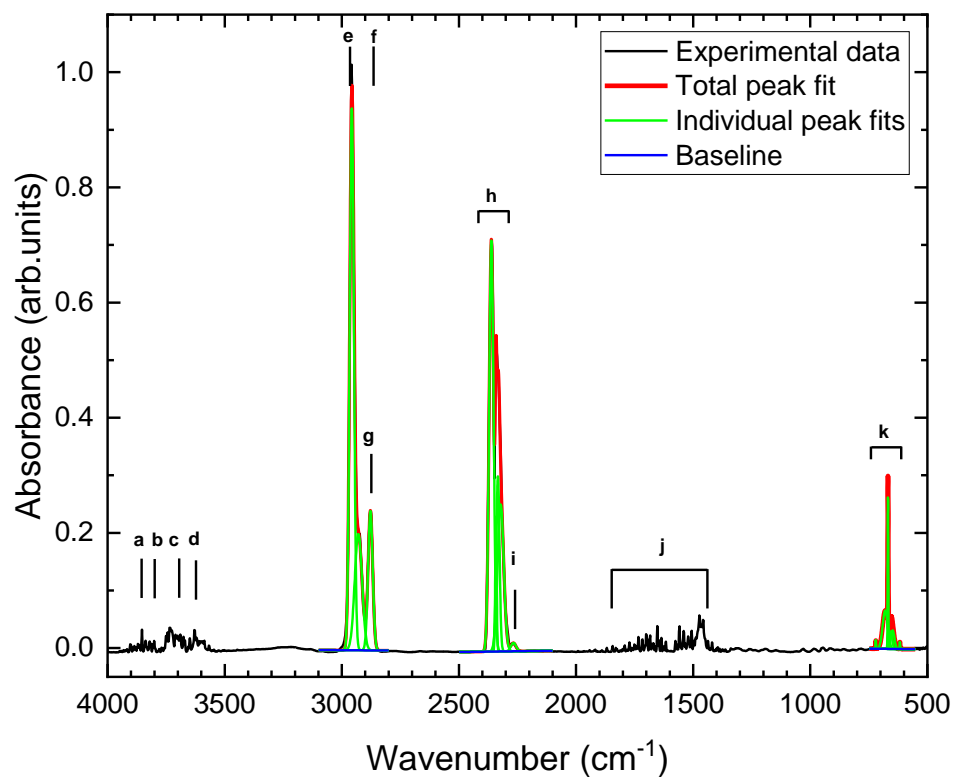
**Figure 5.** Temperature temporal profiles produced by igniting droplets of (a) zeolite-JP-10 and zeolite-JP-10-surfactant and (b) zeolite-Al-JP-10 and zeolite-Al-JP-10-surfactant. The laser irradiation started at  $t = 0$  s.



**Figure 6.** UV-Vis emission spectrum produced during ignition of droplets of (a) zeolite-JP-10 and (b) zeolite-JP-10-surfactant. The total fit (red line) was obtained by simultaneously optimizing a black-body background and Gaussian peaks (green lines) across the 270 to 1100 nm wavelength range. Here, the black-body background of (a)  $2307 \pm 60$  K and (b)  $2223 \pm 70$  K has been subtracted to show the emission peaks and bands more clearly. The assignments of bands a-s are presented in Table 3.



**Figure 7.** UV-Vis emission spectrum produced during ignition of droplets of (a) zeolite-Al-JP-10 and (b) zeolite-Al-JP-10-surfactant. The total fit (red line) was obtained by simultaneously optimizing a black-body background and Gaussian peaks (green lines) across the 270 to 1100 nm wavelength range. Here, the black-body background of (a)  $2225 \pm 70$  K and (b)  $2200 \pm 65$  K has been subtracted to show the emission peaks and bands more clearly. The assignments of bands a-t are presented in Table 4.



**Figure 8.** FTIR transmission spectrum following ignition of droplets of zeolite-JP-10-surfactant. The rovibrational peaks/bands of the products and unreacted JP-10 are labelled (a)–(k) and assigned in Table 5. The total fit (red line) and individual peak fits peaks (green lines) are shown. FTIR transmission spectra of other three systems are shown in supporting information.



**Table 1.** Compilation of prior experimental studies on the pyrolysis of JP-10.

Method	Temperature (K)	Pressure (bar)	Residence time (ms)	Ref.
Vacuum Ultraviolet Photoionization (ALS/NSRL)	1100-1600/ 927-1083	0.8/1.101	A few 0.01/ 124-144	10
Shock tube	1166-1522	2.53-3.04	-	24
Electrically heated tube	823-1008	45	-	25
Flow reactor with fixed bed/ microflow tube	673-733/ 700-980	60/1	6500-13000/ 400	26
Shock tube	1000-1600	6-8	0.5	7
Flow tube reactor	298-1700	0.002-0.004	2.10-9.35	14
Flow tubular reactor	930-1080	1.7	3.2-5.3	8
Jet-stirred reactor	848-933	1	500-6000	16
Annular tubular reactor	903-968			18
System for thermal diagnostic studies	373-873	34	1038-5000	23
System for thermal diagnostic studies	473-935	34	1,800	1
Batch reactor	823-903	1-38	480-26,400	19
Tubular reactor	883-963	1	$1.8 \times 10^6$	20
Thermal block	623-698	345	$2.4 \times 10^5$ $7.2 \times 10^7$	3
Batch reactor	583-683	40	$3.6 \times 10^7$	17
Flow reactor	900-1600	0.00667	-	15

**Table 2.** Physicochemical properties of zeolite samples.

Name	Si/Al ratio <sup>a</sup>	Average crystal size <sup>b</sup> (nm)	$S_{\text{BET}}^{\text{c}}$ ( $\text{m}^2\text{g}^{-1}$ )	$V_{\text{tot}}^{\text{d}}$ ( $\text{cm}^3\text{g}^{-1}$ )	Acidity <sup>e</sup> ( $\text{mmol g}^{-1}$ )		
					Total	Weak	Strong
HZSM-5	65	$218 \pm 22$	339	0.26	0.035	0.020	0.015
Silicalite-1	$\infty$	$216 \pm 18$	308	0.19	0.010	0.006	0.004

<sup>a</sup>Si/Al ratio determined by ICP-OES.

<sup>b</sup>Average crystal size determined by SEM images in Figure 4.

<sup>c</sup> $S_{\text{BET}}$ , BET surface area calculated by BET theory.

<sup>d</sup> $V_{\text{tot}}$ , total pore volume calculated at  $P/P_0 = 0.95$  by BJH theory using adsorption branch of  $\text{N}_2$  adsorption isotherm in Figure 2(a).

<sup>e</sup>Amount of acidic sites determined by  $\text{NH}_3$  TPD-MS in Figure 2(b).

**Table 3.** Vibrational mode assignments for the peaks or bands in the UV-Vis emission spectrum of droplets of zeolite-JP-10 and zeolite-JP-10-surfactant.

Peak or band	Peak wavelength or band center (nm) <sup>a</sup> Zeolite	Peak wavelength or band center (nm) <sup>a</sup> Zeolite-Span80	Molecule, atom or radical	Ref. wavelength (nm)	Transition	Branch; vibrational quantum numbers: (v', v'') or (v <sub>1</sub> ', v <sub>2</sub> ', v <sub>3</sub> ') - (v <sub>1</sub> '', v <sub>2</sub> '', v <sub>3</sub> '')
a	286.5	286.5	OH	287.5-289.3 <sup>b</sup>	A <sup>2</sup> Σ <sup>+</sup> – X <sup>2</sup> Π	R <sub>1</sub> , R <sub>2</sub> , Q <sub>1</sub> , Q <sub>2</sub> ; (2,1)
b	289.5	289.5	OH	287.5-289.3 <sup>b</sup>	A <sup>2</sup> Σ <sup>+</sup> – X <sup>2</sup> Π	R <sub>1</sub> , R <sub>2</sub> , Q <sub>1</sub> , Q <sub>2</sub> ; (2,1)
c	309.5	309.5	OH	309.0 <sup>b</sup>	A <sup>2</sup> Σ <sup>+</sup> - X <sup>2</sup> Π	Q <sub>2</sub> ; (0,0)
d	336.5	336.5	O <sub>2</sub> HCO	337.0 <sup>b</sup> 337.6 <sup>b,i</sup>	B <sup>3</sup> Σ <sub>u</sub> <sup>-</sup> - X <sup>3</sup> Σ <sub>g</sub> <sup>-</sup> Ã <sup>2</sup> A' - X̃ <sup>2</sup> A'	(0,14)
e		354.5	OH	342-359 <sup>b, c</sup>	A <sup>2</sup> Σ <sup>+</sup> - X <sup>2</sup> Π	(1,0), (2,1)
f		389.0	CH	390.0 <sup>b, c</sup>	A <sup>2</sup> Δ – X <sup>2</sup> Π	(0,0)
g	300-580 (broad)	300-580 (broad)	OH, C <sub>2</sub> , CH	Ref.b		
h	589.0	589.0	Na	589.59 <sup>d</sup> 589.00 <sup>d</sup>	<sup>2</sup> S <sub>1/2</sub> - <sup>2</sup> P <sub>3/2</sub> <sup>2</sup> S <sub>1/2</sub> - <sup>2</sup> P <sub>1/2</sub>	
i	613.5	613.0	C <sub>2</sub>	612.2 <sup>b</sup>	d <sup>3</sup> Π <sub>g</sub> – a <sup>3</sup> Π <sub>u</sub>	(1,3)
j	678.1	676.1	H <sub>2</sub> O	632-683 <sup>e</sup>	ro-vib. mode	(1,1,3) - (1,5,1)
k	701.5	701.5	H <sub>2</sub> O	690-710 <sup>f</sup>	ro-vib. mode	(1,0,3) - (0,0,0)
l	753.5	753.5	H <sub>2</sub> O	719-795 <sup>e</sup>	ro-vib. mode	(1,2,2) - (0,1,3)
m	767.5	767.5	K	767.50 <sup>d, g</sup> 769.90 <sup>d, g</sup>	<sup>2</sup> S <sub>1/2</sub> - <sup>2</sup> P <sub>3/2</sub> <sup>2</sup> S <sub>1/2</sub> - <sup>2</sup> P <sub>1/2</sub>	
n	781.0	788.0	H <sub>2</sub> O	719-795 <sup>e</sup>	ro-vib. mode	(1,2,2) - (0,1,3)
o		802.5	H <sub>2</sub> O	719-795 <sup>e</sup>	ro-vib. mode	(1,2,2) - (0,1,3)
p	849.5	849.5	H <sub>2</sub> O	847-906 <sup>e</sup>	ro-vib. mode	(0,0,3) - (1,3,1)
q	892.5	883.5	H <sub>2</sub> O	847-906 <sup>e</sup>	ro-vib. mode	(0,0,3) - (1,3,1)
r	944.5 Opt.fiber.absorp.	944.5 Opt.fiber.absorp.	H <sub>2</sub> O	928-966 <sup>f</sup> Ref. h	ro-vib. mode	(2,0,1) - (0,0,0)
s	998.5	998.5	H <sub>2</sub> O	972-1017 <sup>e, h</sup>	ro-vib. mode	(2,2,0) - (0,4,1)

<sup>a</sup> Measurements are accurate to within 2 nm.

<sup>b</sup> Ref. (59). <sup>c</sup> Ref. (60). <sup>d</sup> Ref. (61). <sup>e</sup> Ref. (64). <sup>f</sup> Ref. (62). <sup>g</sup> Ref. (63). <sup>h</sup> Ref. (46). <sup>i</sup> Ref. (65).

**Table 4.** Vibrational mode assignments for the peaks or bands in the UV-Vis emission spectrum of droplets of zeolite-Al-JP-10 and zeolite-Al-JP-10-surfactant.

Peak or band	Peak wavelength or band center (nm) <sup>a</sup>  Zeolite-Aluminium	Peak wavelength or band center (nm) <sup>a</sup>  Zeolite-Aluminium-Span80	Molecule, atom or radical	Ref. wavelength (nm)	Transition	Branch; vibrational quantum numbers:  (v', v'') or (v <sub>1</sub> ', v <sub>2</sub> ', v <sub>3</sub> ') - (v <sub>1</sub> '', v <sub>2</sub> '', v <sub>3</sub> '')
a		288.5	OH	287.5-289.3 <sup>b</sup>	A <sup>2</sup> Σ <sup>+</sup> – X <sup>2</sup> Π	R <sub>1</sub> , R <sub>2</sub> , Q <sub>1</sub> , Q <sub>2</sub> ; (2,1)
b	309.5	309.5	OH	309.0 <sup>b</sup>	A <sup>2</sup> Σ <sup>+</sup> - X <sup>2</sup> Π	Q <sub>2</sub> ; (0,0)
c		337.1	O <sub>2</sub>  HCO	337.0 <sup>b</sup>  337.6 <sup>b, i</sup>	B <sup>3</sup> Σ <sub>u</sub> <sup>-</sup> -X <sup>3</sup> Σ <sub>g</sub> <sup>-</sup>  Ã <sup>2</sup> A' -X <sup>2</sup> A'	(0,14)
d		355.0	OH	342-359 <sup>b, c</sup>	A <sup>2</sup> Σ <sup>+</sup> - X <sup>2</sup> Π	(1,0), (2,1)
e		388.5	C-H	390.0 <sup>b, c</sup>	A <sup>2</sup> Δ – X <sup>2</sup> Π	(0,0)
f	300-580 (broad)	300-580 (broad)	OH, C <sub>2</sub> , CH	Ref. b		
g	589.5	589.0	Na	589.59 <sup>d</sup>  589.00 <sup>d</sup>	<sup>2</sup> S <sub>1/2</sub> - <sup>2</sup> P <sub>3/2</sub>  <sup>2</sup> S <sub>1/2</sub> - <sup>2</sup> P <sub>1/2</sub>	
h	612.5	613.0	C <sub>2</sub>	612.2 <sup>b</sup>	d <sup>3</sup> Π <sub>g</sub> – a <sup>3</sup> Π <sub>u</sub>	(1,3)
i	642.5		H <sub>2</sub> O	632-683 <sup>e</sup>	ro-vib. mode	(1,1,3) - (1,5,1)
j	678.5	678.1	H <sub>2</sub> O	632-683 <sup>e</sup>	ro-vib. mode	(1,1,3) - (1,5,1)
k	702.5	708.0	H <sub>2</sub> O	690-710 <sup>f</sup>	ro-vib. mode	(1,0,3) - (0,0,0)
l	753.5	752.5	H <sub>2</sub> O	719-795 <sup>e</sup>	ro-vib. mode	(1,2,2) - (0,1,3)
m	769.5	769.5	K	767.50 <sup>d, g</sup>  769.90 <sup>d, g</sup>	<sup>2</sup> S <sub>1/2</sub> - <sup>2</sup> P <sub>3/2</sub>  <sup>2</sup> S <sub>1/2</sub> - <sup>2</sup> P <sub>1/2</sub>	
n	778.0	778.0	H <sub>2</sub> O	719-795 <sup>e</sup>	ro-vib. mode	(1,2,2) - (0,1,3)
o	802.5		H <sub>2</sub> O	719-795 <sup>e</sup>	ro-vib. mode	(1,2,2) - (0,1,3)
p	832.5		H <sub>2</sub> O	811-839 <sup>e</sup>	ro-vib. mode	(2,1,1) - (0,0,0)
q	847.5	847.5	H <sub>2</sub> O	847-906 <sup>e</sup>	ro-vib. mode	(0,0,3) - (1,3,1)
r	888.5	888.5	H <sub>2</sub> O	847-906 <sup>e</sup>	ro-vib. mode	(0,0,3) - (1,3,1)

s	944.5 Opt.fiber.absorp	944.5 Opt.fiber.absorp.	H <sub>2</sub> O	928-966 <sup>f</sup> Ref. h	ro-vib. mode	(2,0,1) - (0,0,0)
t	998.5	998.5	H <sub>2</sub> O	972-1017 <sup>e, h</sup>	ro-vib. mode	(2,2,0) - (0,4,1)

<sup>a</sup> Measurements are accurate to within 2 nm.

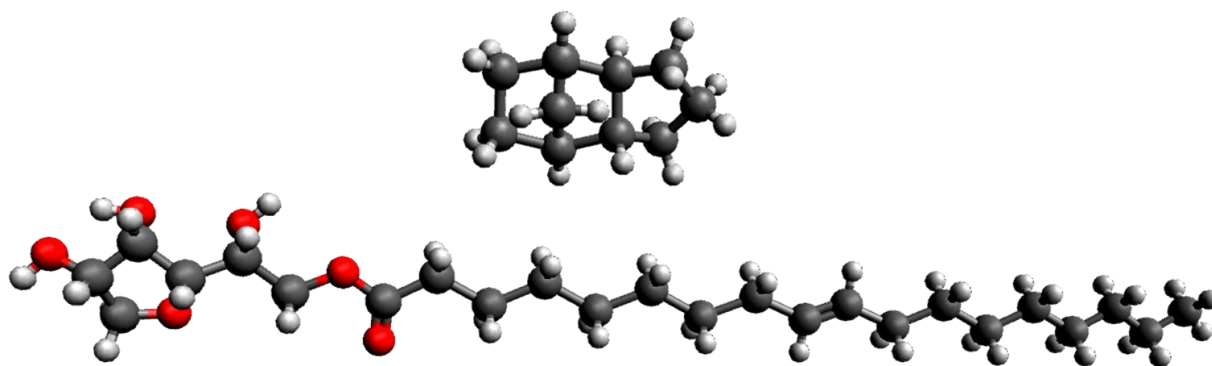
<sup>b</sup> Ref. (59). <sup>c</sup> Ref. (60). <sup>d</sup> Ref. (61). <sup>e</sup> Ref. (64). <sup>f</sup> Ref. (62). <sup>g</sup> Ref. (63). <sup>h</sup> Ref. (46). <sup>i</sup> Ref. (65).

**Table 5.** Vibrational mode assignments for the peaks or bands in the FTIR transmission spectrum of droplets of zeolite-JP-10.

Band	Peak or band wavenumber <sup>a</sup> (cm <sup>-1</sup> )	Molecule	Ref. band (cm <sup>-1</sup> )	Number (symmetry)	vibrational mode <sup>b</sup>
a	3756	H <sub>2</sub> O	3756 <sup>c</sup>	3(b <sub>1</sub> )	$\nu_{\text{as}}(\text{H}_2\text{O})$
b	3714	CO <sub>2</sub>	3714.8 <sup>d</sup>	1( $\sigma_{\text{g}}^+$ ) + 3( $\sigma_{\text{u}}^+$ )	combination band
c	3657	H <sub>2</sub> O	3657 <sup>c</sup>	1(a <sub>1</sub> )	$\nu(\text{H}_2\text{O})$
d	3612	CO <sub>2</sub>	3612.8 <sup>d</sup>	1( $\sigma_{\text{g}}^+$ ) + 3( $\sigma_{\text{u}}^+$ )	combination band
e	2956	C <sub>10</sub> H <sub>16</sub>	2942 <sup>e</sup>	68	$\nu(\text{CH})$
f	2929	C <sub>10</sub> H <sub>16</sub>	2913 <sup>e</sup>	62	$\nu(\text{CH}_2)$
g	2879	C <sub>10</sub> H <sub>16</sub>	2865 <sup>e</sup>	57	$\nu(\text{CH})$
h	2347	CO <sub>2</sub>	2349.1 <sup>d</sup>	3 ( $\sigma_{\text{u}}^+$ )	$\nu_{\text{as}}(\text{CO}_2)$
	2359	CO <sub>2</sub>	2300 – 2380 <sup>f</sup>		$\nu_{\text{as}}(\text{CO}_2)$
i	2279	<sup>13</sup> C <sup>16</sup> O <sub>2</sub>	2283 <sup>c</sup>	3 ( $\sigma_{\text{u}}^+$ )	$\nu_{\text{as}}(\text{CO}_2)$
j	1600	H <sub>2</sub> O	1595 <sup>c</sup>	2(a <sub>1</sub> )	$\delta(\text{OH})$
	1457	C <sub>10</sub> H <sub>16</sub>	1456 <sup>e</sup>	54	scissoring (CH <sub>2</sub> )
	1466	C <sub>10</sub> H <sub>16</sub>	1468 <sup>e</sup>	55	scissoring (CH <sub>2</sub> )
k	668	CO <sub>2</sub>	667.4 <sup>d</sup>	2( $\pi_{\text{u}}$ )	$\delta(\text{CO}_2)$
	648	<sup>13</sup> C <sup>16</sup> O <sub>2</sub>	649.0 <sup>c</sup>	2( $\pi_{\text{u}}$ )	$\delta(\text{CO}_2)$
	619, 655, 680, 720	CO <sub>2</sub>	620-710 <sup>f</sup>	2( $\pi_{\text{u}}$ )	$\delta(\text{CO}_2)$

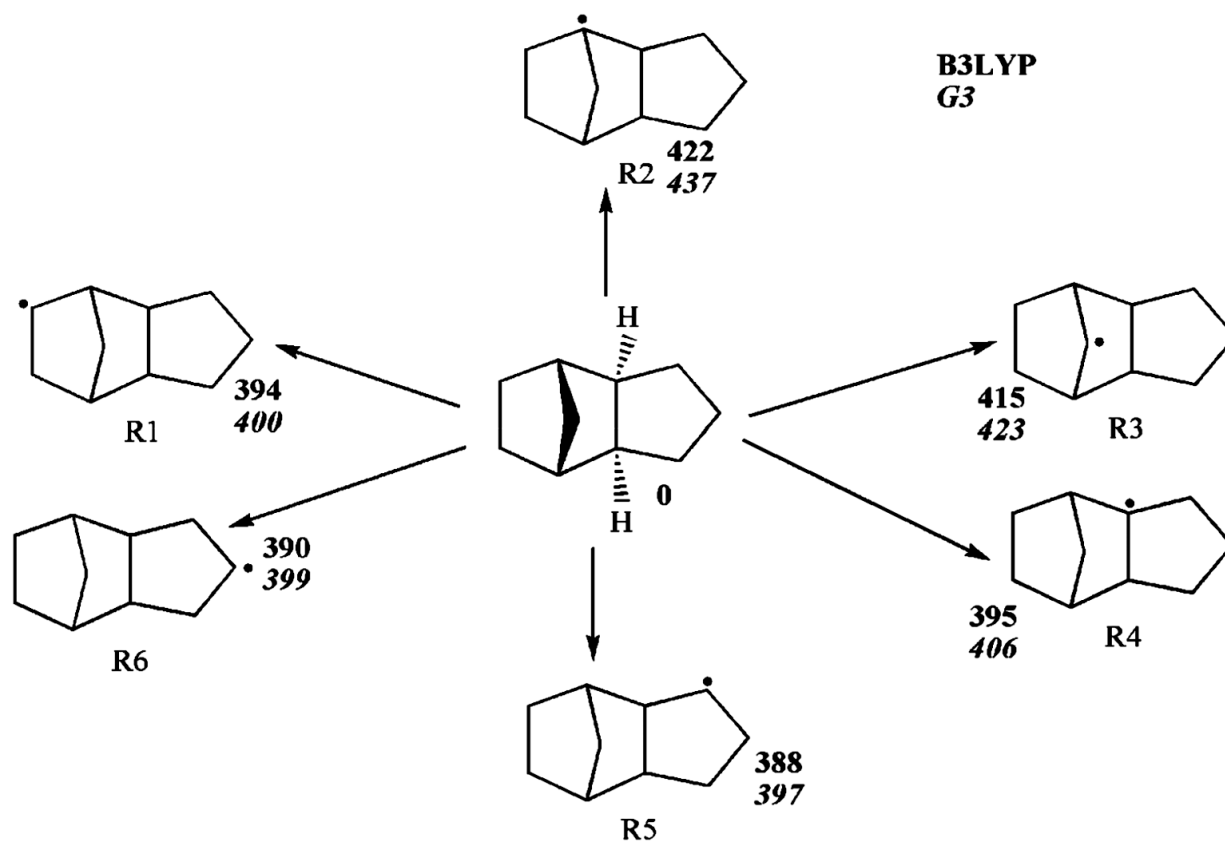
<sup>a</sup> Spectral resolution of 4 cm<sup>-1</sup>.

<sup>b</sup>  $\nu$  denotes stretch; bend; and as, antisymmetric. <sup>c</sup> Ref. (61). <sup>d</sup> Ref. (66). <sup>e</sup> Ref. (47). <sup>f</sup> Ref. (67).



**Scheme 1.** Molecular structure of exo-tetrahydrodicyclopentadiene (JP-10) (top) and the surfactant (bottom). Carbon, hydrogen, and oxygen atoms are color coded in grey, white, and red, respectively.





**Scheme 2.** Radicals generated via C-H bond cleavages in JP-10.<sup>10</sup> The reaction energies for the C-H bond ruptures were calculated at the B3LYP/6-311G\*\* and G3 level and are given in kJ mol<sup>-1</sup>.