The synergistic dispersion and biodegradation for oil in seawater

based on Janus nanosheets and oil-degrading bacteria

Haosen Zhao^{*a*, *b*}, Xin Hu^{*a*, *b*}, Hu Kang^{*a*, *b*}, Dongdong Yang^{*a*, *b*}, Mutai Bao^{*a*, *b*}, Yiming

Li ^{*a, b* *}

^{*a*} Frontiers Science Center for Deep Ocean Multispheres and Earth System/Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, 266100, Qingdao, P.R. China

^b College of Chemistry and Chemical Engineering, Ocean University of China, 266100,

Qingdao, P.R. China

^{*} Corresponding authors.

E-mail address: liym@ouc.edu.cn (Y. M. Li).

Text 1. The preparation of HSMA

150 mL toluene, 5.9220 mL styrene, 5.0516 g maleic anhydride, 0.1042 g 2, 2-Azobis(2-methylpropionitrile) were added in 250 mL three-mouth flask with stirring. After purging with nitrogen for 30 min, the mixturewas heated to 85°C and then refluxed for 3 h to obtain a white emulsion. A certain amount of toluene was added to wash the white product and dried in a vacuum oven at 60°C.

5.0 g of the above products were dissolved in 45 g of 3 wt% sodium hydroxide solution and stirred at 80°C for 3 hours to get 10 wt% hydrolyzed styrene-maleic anhydride (10 wt% HSMA).

Text 2. Silica particles as markers to load on the JNs

2.0 g of JNs, 1 ml of TEOS and 10ml of water were added to a beaker and stirred at 70°C for 12 hours, then washed with water and dried in a vacuum oven at 60 °C.

Text 3. Immobilization of bacteria

2ml of bacterial suspension was absorbed and centrifuged at 1000 r · min⁻¹ for 2min, and washed three times with sterile water. 2.5% glutaraldehyde was added to the bacterial seed solution and then stood for 30min.

Gradient dehydration method was performed by ethanol: 75%, 90% and absolute ethanol were successively allowed to stand for 15min, and then centrifuged. Finally, the immobilized bacteria are dried for 3 hours at 85 ° C.



Fig. S1 Schematic illustration of (a) JNs preparation and (b) the process of JNs combining with AP-1 bacteria.



Fig. S2 XPS spectrum of Si2p.

Name	Materials	Oil phase	Material	Oil-water	Oil droplet	References
			concentration	volume ratio	size	
Carboxyl- terminated carbon black	carbon black	BP-MC 252 crude oil	0.015 wt %	1:9	~150 µm	6
Kaolinite	natural clay	Anadarko crude oil	25 g/L	1:30	~150 µm	9
Janus silica particles (JSPs)	silica	n-tetradecane	12 g/L	1:4	$\sim 50 \ \mu m$	14
Polymer- inorganic Janus nanosheets	silica	toluene	1.0 wt%	1:2	$\sim 100 \ \mu m$	18
Poly (allylamine hydrochloride)- attapulgite	attapulgite	n-tetradecane	20 g/L	1:4	~33 µm	31
Janus nanosheets	silica	diesel oil	8 g/L	1:4	$\sim 50~\mu m$	This work

1 1 1



Fig. S3 (a) Emulsification of JNs, (b) Synergistic emulsification of JNs and bacteria $(OD_{600}=1.72)$. Both groups were emulsified under the following conditions: concentration of JNs is 0.08 g/L, the volume ratios of diesel-seawater is 1:4, stabilization time is 24 h.



Fig. S4 The transmission profiles of (a) different concentrations of JNs, (b) different concentrations of AP-1. Both groups were stable under the following conditions: the number of spectral lines is 300, the time interval is 1 s, the speed is 1500 rpm, the light intensity is 1, and the light source is 870 nm.



Fig. S5 The curve of OD value and bacterial concentration.



Fig. S6 The number of colonies at different concentrations of JNs (a) 0 g/L, (b) 0.2 g/L, (c) 0.4 g/L, (d) 0.6 g/L, (e) 0.8 g/L.



Fig.S7 The biodegradation and fitting curves of diesel in the presence and absence of JNs respectively.

The data of degradation at different times was normalized according to Fig. 8 (c), as shown in Table 1, and then fitted the processed data according to the first-order dynamic equations (3) and second-order dynamic equations (4):

$$lnC = -k_1 t + lnC_0 \qquad t_1 = \frac{ln2}{k_1} \qquad (3)$$

$$\frac{1}{C} = k_2 t + \frac{1}{C_0} \qquad t_1 = \frac{1}{k_2 C_0}$$
(4)

-

t	0	1	2	3	4	5	6	7
С	59.397	51.123	42.810	36.001	28.051	22.225	19.208	16.637
C'	59.397	39.946	21.969	15.765	13.682	9.356	8.605	8.394
lnC	4.084	3.934	3.757	3.584	3.334	3.101	2.955	2.812
1/Cʻ	0.017	0.025	0.046	0.063	0.073	0.107	0.116	0.119

Table S2 Processed gas chromatography data

where, t is the time, C is the diesel concentration and C_0 is the initial diesel concentration. The concentrations of targeted compounds/compound groups were normalized against hopene.