Petroleum Science 18 (2021) 1543–1550

Contents lists available at ScienceDirect

Petroleum Science

journal homepage: www.keaipublishing.com/en/journals/petroleum-science

Original Paper

Systematic adsorption process of petroleum hydrocarbon by immobilised petroleum-degradation bacteria system in degradation pathways

Xin-Ge Fu [a], Ke Shi [a], Jian-Liang Xue [a, *], Chuan Chen [b], Yu Bai [c], Yan-Lu Qiao [a], Yi-Xuan Liu [a], Xiang-Ming Hu [a], Yu Gao [a], Hao Yu [d]

[a] College of Safety and Environmental Engineering, Shandong University of Science and Technology, Qingdao, Shandong, 266590, China
[b] School of Environment, Harbin Institute of Technology, Haerbin, Heilongjiang, 150001, China
[c] Chinaunicom System Integration Co, Ltd, No.131, Xidan North Road, Beijing, 100085, China
[d] College of Chemical and Biological Engineering, Shandong University of Science and Technology, Qingdao, Shandong, 266590, China

ARTICLE INFO

Article history:
Received 2 October 2020
Accepted 16 April 2021
Available online 2 September 2021

Edited by Xiu-Qiu Peng

Keywords:
Petroleum hydrocarbon pollution
Immobilisation
Adsorption-biodegradation
Molecular simulation
Bioremediation

ABSTRACT

In order to effectively control petroleum hydrocarbon pollution by immobilisation technology, it is necessary to understanding the degradation pathways of petroleum hydrocarbon in immobilised petroleum-degradation bacteria system. However, the adsorption in degradation process is rarely studied. In this study, adsorption process of petroleum hydrocarbon was synergistically studied by means of the surface properties, adsorption thermodynamics and molecular simulation. The results indicate that the immobilised petroleum-degradation bacteria have many holes for the bacteria to adsorb. The diesel adsorption by immobilised petroleum-degradation bacteria is a spontaneous, entropy-increasing and endothermic process. Diesel is first adsorbed to the surface of immobilised petroleum-degradation bacteria through hydrogen bonding, and then is biodegraded. This study provided substantial knowledge of immobilised technology in controlling petroleum hydrocarbon pollution.

1. Introduction

Offshore petroleum transportation is increasingly developing, and petroleum hydrocarbon leakage accidents occur frequently (Li et al., 2009; Liu et al., 2020; Partovinia and Rasekh, 2018). This petroleum hydrocarbon pollution causes huge economic and environmental losses (Geng et al., 2013; Helle et al., 2015; Tang et al., 2019). The establishment of practical and effective restoration and control measures is imminent. Bioremediation has become an important means to solve petroleum hydrocarbon pollution due to its low cost, environment friendly and thorough degradation characteristics (Alabresm et al., 2018; Du et al., 2011; Marin et al., 2005; Wu et al., 2019). However, bioremediation technology has many limitations, such as small microbial density in practical application (Head et al., 2006; Pi et al., 2019). With the deepening of research, scholars have found a mothod called immobilised microorganism technology to effectively solve the above problems (Xue et al., 2017; Li et al., 2019b).

Immobilised microorganism technology is a bioremediation technology where free microorganisms are immobilised on specific carriers by using physical or chemical methods. A series of studies showed that it is an important method to treat petroleum hydrocarbon contamination (Fu et al., 2021; Li et al., 2019a). Many scholars have prepared materials with immobilised petroleum-degradation bacteria with high degradation rate (Kumar et al., 2019). The bacteria immobilised in a modified bamboo charcoal (94%) have a more efficient diesel degradation efficiency than free bacteria (82%) (Chen et al., 2016). Xue et al. (2019) prepared immobilised bacteria in straw-alginate beads by utilising straw as a carrier. The diesel degradation rate by immobilised petroleum-degradation bacteria in 5 days is 68.68%, which is larger than that of free cell (53.32%). The diesel degradation rate by immobilised petroleum-degradation bacteria is higher than free bacteria. This condition is extremely necessary to treat petroleum hydrocarbon pollution (Garcia et al., 2019; Zhang et al., 2018).

Extensive studies have been widely conducted on petroleum

* Corresponding author.
E-mail address: ll-1382@163.com (J.-L. Xue).

https://doi.org/10.1016/j.petsci.2021.09.009

hydrocarbon removal by immobilised petroleum-degradation bacteria (Chandran and Das, 2011; Li et al., 2011). As early as 1980, some scholars used numerical equations to predict biodegradation process (Geng et al., 2014). Most of the analysis focused on the limiting factors of biodegradation, such as, salinity, carrier materials and temperature (Niu et al., 2013). However, few studies have explored the combination modes of petroleum hydrocarbon and components in the immobilised petroleum-degradation bacteria and the degradation pathways of petroleum hydrocarbon in the immobilised petroleum-degradation bacteria system. In particular, the transfer of petroleum hydrocarbon and the degradation inside and outside of the immobilised petroleum-degradation bacteria are ignored. Many phases, such as adsorption of diesel and biodegradation of diesel, are found in the degradation of immobilised petroleum-degradation bacteria. Among them, adsorption plays an important role in the degradation of petroleum hydrocarbons. However, the adsorption process is rarely studied.

To systematically study the adsorption process, the surface properties of material, adsorption thermodynamics and the combination modes of petroleum hydrocarbon with material components based on molecular simulation were studied. The study provides technical support and theoretical basis for the bioremediation of petroleum hydrocarbon pollution.

2. Material and methods

2.1. Chemicals

0# diesel was obtained from the 151th Station in Qingdao, Shandong Province and filtered with a membrane (0.22 μm) prior to use. The cinnamon shells were ground in 100 mesh powder with a crusher. Other chemicals were obtained from Qingdao Jingke Instrument Reagent Co., Ltd.

2.2. Preparation of immobilised petroleum-degradation bacteria

In this study, the YT-11 strain (*Pseudomonas* sp.) was gained from the seawater. The cinnamon shell was the best immobilised carrier obtained through screening due to the excellent degradation performance of the immobilised petroleum-degradation bacteria prepared with cinnamon shell as carrier (degradation rate in 5 days is 69.57%) (Fu et al., 2019). The immobilised petroleum-degradation bacteria in this study are composed of sodium alginate (SA), cinnamon shell powder, *Pseudomonas* sp. YT-11 and $CaCl_2$. They appear as small particles of 2–3 mm.

SA is a widely used water-soluble alginate. SA can quickly undergo ion exchange and form a gel when it meets calcium ions. With this property, alginate colloidal spheres were prepared by dropping alginate solution into aqueous solution containing calcium ions.

$$2AlgNa + Ca^{2+} = (Alg)_2Ca + 2Na^+ \quad (1)$$

The carrier (cinnamon shell) with superior performance obtained from the previous screening was cleaned (Fu et al., 2019). The carrier was immersed in distill water for 5–6 h to remove impurities and then carefully cleaned with alcohol and water for 3 times. Washed carrier was crushed to obtain particulate matter for use (Kim and Park, 2007). Firstly, SA and $CaCl_2$ solutions were prepared. SA, bacterial solution, and carrier were then mixed evenly in proportion. Finally, the mixed solution was dropped into the $CaCl_2$ solution to form immobilised petroleum-degradation bacteria. The immobilised petroleum-degradation bacteria were removed and washed with physiological saline for two or three times after cross-linking at 4 °C for 36 h.

2.3. Scanning electron microscopy (SEM) analysis

The morphology of immobilised petroleum-degradation bacteria before and after degradation was observed through SEM (APREO, FEI, USA). Glutaraldehyde solution (2.5%) and PBS (0.2 M) were prepared in advance. The immobilised petroleum-degradation bacteria were fixed in glutaraldehyde solution (2.5%) at 4 °C for 4 h, and then rinsed in PBS for 3 times, each time for 10 min. These bacteria were dehydrated with ethanol solution of different concentrations (30%, 50%, 70%, 90% and 100%), and each concentration was balanced for 10 min. The specimens were dried with a freeze dryer, treated with gold spray, fixed with silver glue, and observed through SEM.

2.4. Degradation kinetics

The biodegradation rate equation of organics has always been a hot topic for researchers. Comprehensively understanding and analysing the degradation by studying the degradation rate and reaction order of organics are valuable (Zahed et al., 2011). The research shows that an exponential rate model can simulate the organic biodegradation in a wide range. The relationship between the degradation rate of exponential rate model and matrix mass fraction is expressed as follows:

$$-ds/dt = KS^n \quad (2)$$

where S (mg/L) is the matrix mass fraction, K is the rate constant, t (d) is the reaction time, n is the reaction series.

For the zero-order, the first-order and second-order reactions, the integral form of the above formula is as follows:

$$n = 0, S_0 - S_t = Kt \quad (3)$$

$$n = 1, \ln S_0/S_t = Kt \quad (4)$$

$$n = 2, l/S_t - 1/S_0 = Kt \quad (5)$$

where S_t (mg/L) represents the concentration of matrix at time t, S_0 (mg/L) represents the mass concentration of matrix at initial time, t (d) represents the reaction time, and K represents the rate constant.

In this study, put the immobilised petroleum-degradation bacteria into 100 mL of mineral salt medium (Fu et al., 2020). The initial concentrations of diesel and immobilised petroleum-degradation bacteria are 7.5 g/L. The above solution was cultured in a shaker (120 rpm/min, 29 °C). The residual diesel concentration was determined every 12 h. The kinetic equation of diesel degradation by immobilised petroleum-degradation bacteria was established. Each data was the average of three independent repeated experiments (±SD).

2.5. Adsorption study

The immobilised petroleum-degradation bacteria (1.5 g) were accurately weighed and placed in 20 mL diesel solution with different concentrations (2–15 g/L). The diesel solution was shaken in a constant temperature shaker (120 rpm/min) under different temperature conditions for 3 h. The concentration of diesel was measured, and isothermal equilibrium adsorption amount of diesel was calculated (Shi et al., 2021; Xiao et al., 2019; Zhou et al., 2019).

Each data was the average of three independent repeated experiments (±SD).

2.5.1. Adsorption isotherm

Langmuir and Freundlich models are utilised widely to investigate adsorption behaviour. Langmuir isotherm model is utilised to explain monolayer adsorption and can be written as follows:

$$C_e / q_e = 1/(q_m K_L) + C_e/q_m \quad (6)$$

where q_e (mg/g) represents the amount of diesel adsorbed each unit of immobilised petroleum-degradation bacteria, C_e (mg/L) represents the residual diesel concentration at equilibrium, q_m represents the maximum quantity of diesel adsorbed per unit mass of immobilised petroleum-degradation bacteria, and K_L ($L \cdot mg^{-1}$) represents the Langmuir constant.

Freundlich isotherm model simulates the adsorption of multi molecular layers on multilayered heterogeneous surfaces without considering adsorption saturation and can be expressed as follows:

$$\ln(q_e) = \ln(K_F) + \ln(C_e)/n \quad (7)$$

where K_F (mg/g) presents a constant reflecting adsorption capacity, n presents a constant reflecting adsorption strength, and the value of $1/n$ can be used to judge the difficulty of the adsorption process, that is, irreversible adsorption ($1/n = 0$), favourable adsorption ($0 < 1/n < 1$), and unfavourable adsorption ($1/n > 1$).

2.5.2. Adsorption thermodynamics

The thermodynamic parameters of diesel adsorption by immobilised petroleum-degradation bacteria were calculated by utilising the following formulas.

$$\ln(q_e / C_e) = \Delta S/R - \Delta H/RT \quad (8)$$

$$\Delta G = \Delta H - T\Delta S \quad (9)$$

where R (8.314 $J\ mol^{-1}\ K^{-1}$) represents gas constant, ΔH ($kJ \cdot mol^{-1}$), ΔS ($J \cdot mol^{-1}\ K^{-1}$) and ΔG ($kJ \cdot mol^{-1}$) is the variety in enthalpy, entropy and free energy, respectively. T (K) represents temperature. ΔS and ΔH are gained from the intercepts and slopes of fitting curve of $ln(q_e/C_e)$ versus $1/T$, respectively, and ΔG is obtained from Eq. (9).

2.6. Pathways to remove diesel by immobilised petroleum-degradation bacteria

The adsorption and biodegradation of degraded diesel were measured at different times in accordance with previous methods (Fu et al., 2020). The surface adsorption amount was determined as follows. Firstly, the immobilised petroleum-degradation bacteria were cleaned with inorganic salt solution once. Secondly, they were cleaned twice with extractant and were cleaned twice with inorganic salt solution. The above supernatants were mixed, and diesel adsorption amount was determined through ultraviolet spectrophotometry.

The calculation formula of diesel degradation rate is as follows (Xue et al., 2019).

$$D_t = (C - C_t)/C \times 100\% \quad (10)$$

where D_t (%) represents diesel degradation rate, C (mL/L) represents original diesel concentration, and C_t (mL/L) represents diesel concentration at time t.

2.7. Molecular docking investigations

Molecular Operating Environment software is used to dock small molecules and proteins and explore docking modes. The dehydrogenase produced by bacteria plays an important part in diesel degradation. Apart from a large number of hydrocarbon compounds, diesel has several nonhydrocarbon substances, such as fatty acid, naphthenic acid, thiol and other small molecular substances. In this study, the combination modes of bacteria, diesel and immobilised carrier cinnamon shell were explored. The small molecular structure can be downloaded from http://zinc.docking.org/, and the dehydrogenase and cinnamon shell can be downloaded from http://www.rcsb.org/(Shi et al., 2020).

3. Results and discussion

3.1. Adsorption and biodegradation performance of immobilised petroleum-degradation bacteria

3.1.1. Degradation kinetics

The reaction rate can predict the speed of the reaction approaching equilibrium or quasi equilibrium state, and the experimental rate constant and reaction order can be obtained to reveal the reaction mechanism. The residual diesel concentration data at different times were fitted to the exponential rate equation. The parameters gained are shown in Table 1. Degradation of diesel by immobilised petroleum-degradation bacteria accorded with the second-order reaction well. The correlation coefficients R^2 is 0.99, and the K value is 0.00004 $mg^{-1}\ L \cdot d^{-1}$. Thus, the reaction rate was positively correlated with the square of the diesel concentration. Similarly, the TPH degradation conformed to second-order kinetic equation (Onwosi et al., 2017). Other scholars have found different petroleum hydrocarbon degradation processes. Xue et al. (2019) researched the degradation kinetics of SA-straw microspheres. The results illustrate that the degradation of petroleum hydrocarbon by SA-straw microspheres corresponds to the Monod equation. The K_s and υ_{max} of SA-straw microspheres are 3.23 g/L and 1.84 d^{-1}, respectively. Some scholars found that the degradation kinetics of immobilised *micrococcus* sp. accords with a first-order kinetic model (Hu and Yang, 2015). Microorganisms can regulate their own physiological and biochemical characteristics with different environmental systems. This condition makes it impossible to obtain a unified kinetic equation for the degradation of petroleum hydrocarbon pollutants by microorganisms. Therefore, different kinetic models are shown in these studies.

3.1.2. Different between adsorption and biodegradation of diesel by immobilised petroleum-degradation bacteria

The diesel removal rate in different phases are shown in Fig. 1. Adsorption played an important part in the early stage. On the 3h, the proportion of the adsorbed diesel by immobilised petroleum-degradation bacteria reached above 90%. This condition indicated that the immobilised petroleum-degradation bacteria were an excellent adsorbent in the initial stage. The proportion of biodegradation reached more than 90% on the fifth day. Thus, the degradation of diesel by immobilised petroleum-degradation bacteria depended on adsorption in the initial stage and biodegradation in the later stage. In the degradation process, the immobilised carrier cinnamon shell promoted the growth of bacterial cells and adsorbed diesel into the pores of immobilised petroleum-degradation

Table 1
Parameters of different order reaction equations.

	K	R^2
zero-order reaction	1000, $mg \cdot L^{-1} \cdot d^{-1}$	0.97
first-order reaction	0.1947, d^{-1}	0.98
second-order reaction	0.00004, $mg^{-1} \cdot L \cdot d^{-1}$	0.99

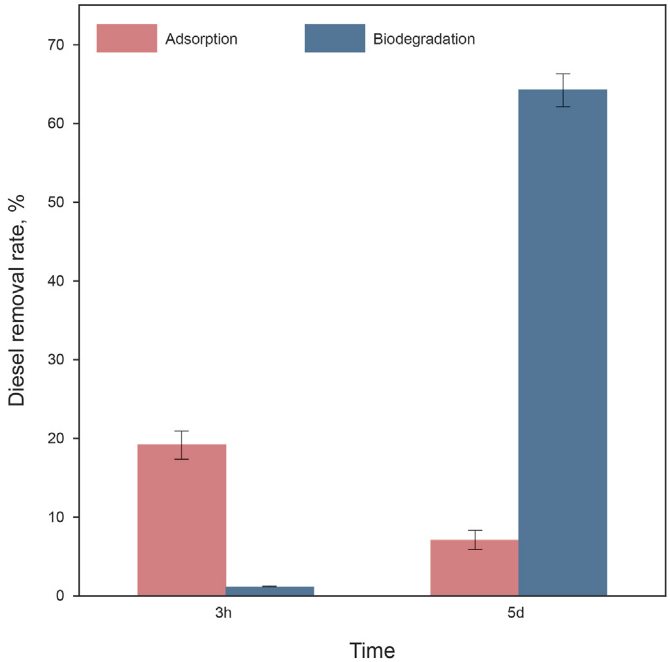

Fig. 1. Diesel removed by immobilised petroleum-degradation bacteria.

bacteria, thereby facilitating the diffusion of diesel. At the same time, the direct contact between diesel and immobilised petroleum-degradation bacteria is conducive to the later biodegradation. Similarly, the strains immobilised on multifunctional materials prepared by Wang et al. (2015) experienced adsorption and biodegradation for diesel removal. Adsorption was a main process in the first stage, and biodegradation was dominant.

Overall, adsorption does play an important role in the process of diesel degradation. In the follow-up of this study, the adsorption process of diesel by immobilised petroleum-degradation bacteria was mainly studied.

3.2. Systematic study on adsorption process

3.2.1. SEM imaging of immobilised petroleum-degradation bacteria

The SEM results of the surface and interior of the immobilised petroleum-degradation bacteria are shown in Fig. 2. The surface of immobilised petroleum-degradation bacteria was loaded with many petroleum-degradation bacteria. The interior of the immobilised petroleum-degradation bacteria had dense and irregularly arranged holes, which was a porous structure. Numerous pores were found for the bacteria to adsorb. This condition can ensure that the immobilised petroleum-degradation bacteria have rich biomass. The interior of the immobilised petroleum-degradation bacteria was rough and porous with wrinkles, which was a good substrate for the bacteria to be fixed. The porous structure was similar to previous observations by Lou et al. (2019). The immobilised petroleum-degradation bacteria were porous.

3.2.2. Adsorption isotherm

Under different temperature conditions, the isothermal adsorption curve of immobilised petroleum-degradation bacteria for diesel is shown in Fig. 3. Adsorption amount increased with enhancement in temperature, revealing that adsorption of diesel is an endothermic process. The fitting parameters of two kinds of isotherms were gained (Table 2) by processing the data of adsorption capacity and equilibrium concentration. Compared with the Freundlich adsorption isotherm, the Langmuir adsorption isotherm has a higher correlation coefficient. The values of $n>1$ suggest that adsorption is easy to proceed.

3.2.3. Adsorption thermodynamics

The fitting curve of $ln(q_e/C_e)$ versus $1/T$ is shown in Fig. 4. Results of thermodynamic experiments are given in Table 3. The result illustrated that ΔG was negative under different temperature conditions, demonstrating that the adsorption was spontaneous. Its absolute value increased with the increase in temperature, suggesting that diesel adsorption is better under a high-temperature condition. The adsorption heat ΔH was positive, revealing that diesel adsorption is an endothermic process. This finding was consistent with the isothermal adsorption result. A positive value of ΔS showed that the free degree of solid–liquid interface increased after diesel adsorption by immobilised petroleum-degradation bacteria. Consequently, the adsorption was a spontaneous, entropy-increasing and endothermic process.

3.2.4. Molecular simulation of bacteria-diesel-carriers

In this study, some diesel compositions and diesel oxygen-containing supplements were used to simulate docking method of cinnamon shell and dehydrogenase with them (Liang et al., 2005; Shi et al., 2020). The results showed that the cinnamon shell or dehydrogenases and some small molecules were bound by hydrogen bonds or weak bonds on the basis of similar compatibility

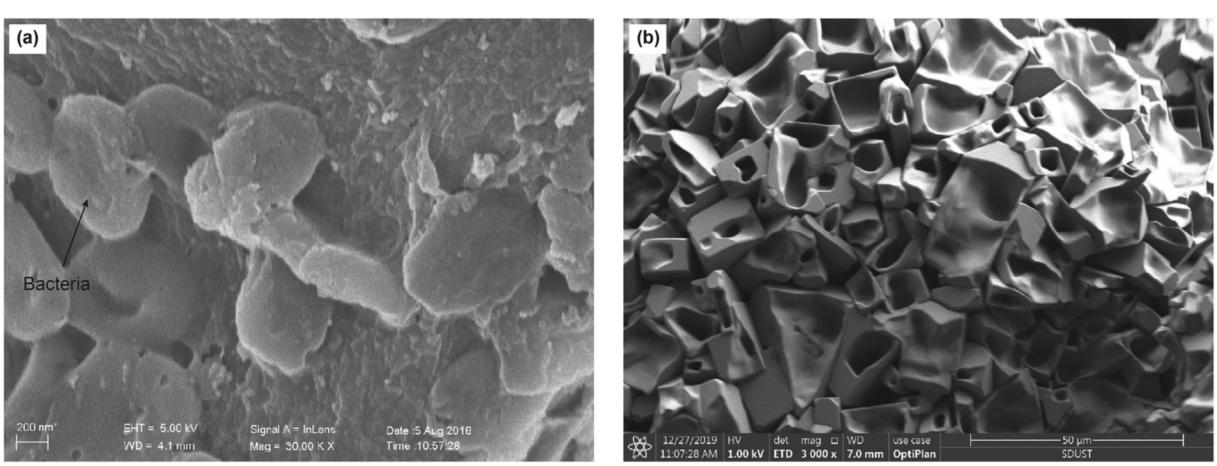

Fig. 2. SEM results of the surface (a) and interior (b) of immobilised petroleum-degradation bacteria.

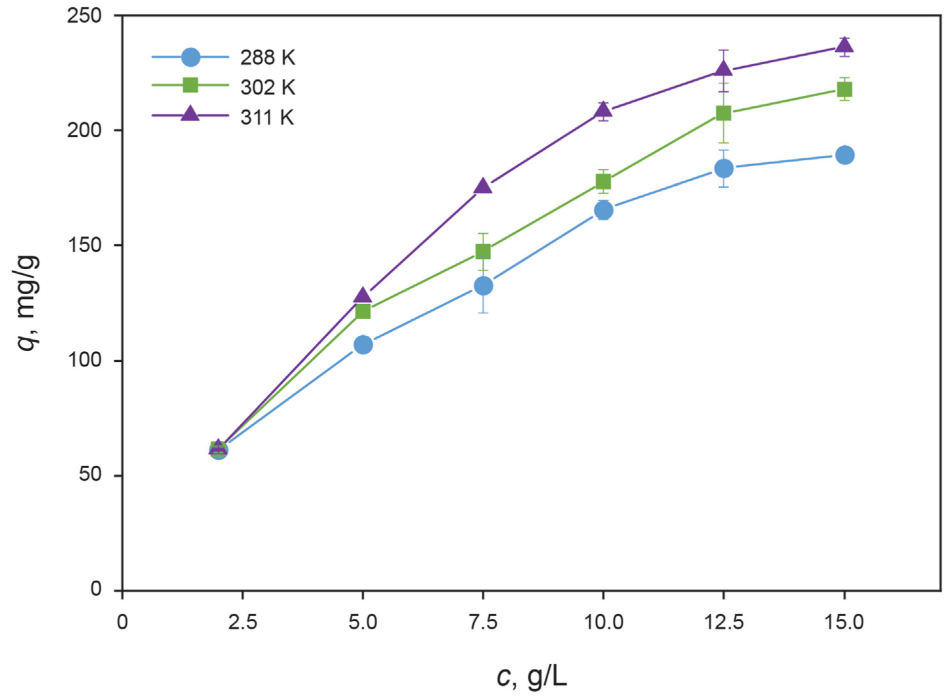

Fig. 3. Isotherm adsorption curve of diesel by immobilised petroleum-degradation bacteria.

Table 2
Fitting parameters of adsorption isotherm model.

T/K	Langmuir adsorption isotherm			Freundlich adsorption isotherm		
	q_m, g/L	K_L, L·mg^{-1}	R^2	K_F, mg/g	n	R^2
288	208.33	0.00064	0.94	21.2	4.26	0.89
302	270.27	0.00025	0.98	22.7	4.13	0.95
311	277.78	0.00088	0.99	27.8	4.60	0.88

Table 3
Parameters for the adsorption thermodynamic of diesel by immobilised petroleum-degradation bacteria.

T, K	ΔG, kJ·mol^{-1}	ΔS, J·mol^{-1}·K^{-1}	ΔH, kJ·mol^{-1}
288	−0.05	30.30	8.68
302	−0.47		
311	−0.74		

principles. Fig. 5 shows the hydrogen bonding mode of a small molecule (benzoic acid) to cinnamon shell and dehydrogenase. The optimal binding mode and binding site were obtained. Fig. 5a and b shows the minimum energy docking structure of benzoic acid with cinnamon shell and dehydrogenase. As shown in Fig. 5a, b, a large pocket in the cinnamon shell or dehydrogenase provided an entrance for benzoic acid to be close to the active center. The logical complex structures are revealed in Fig. 5c and d. In accordance with the molecular simulation results, the cinnamon shell and benzoic

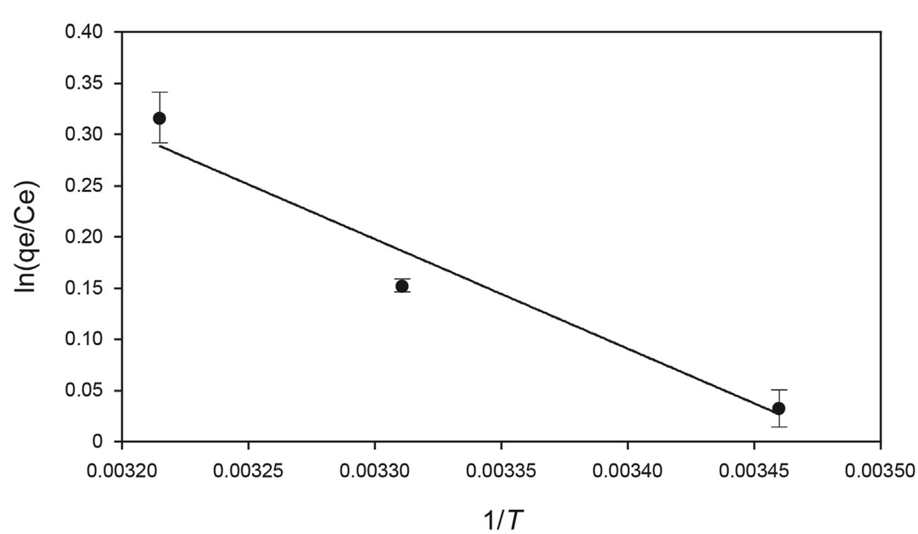

Fig. 4. Fitting curve of ln (q_e/C_e) versus $1/T$.

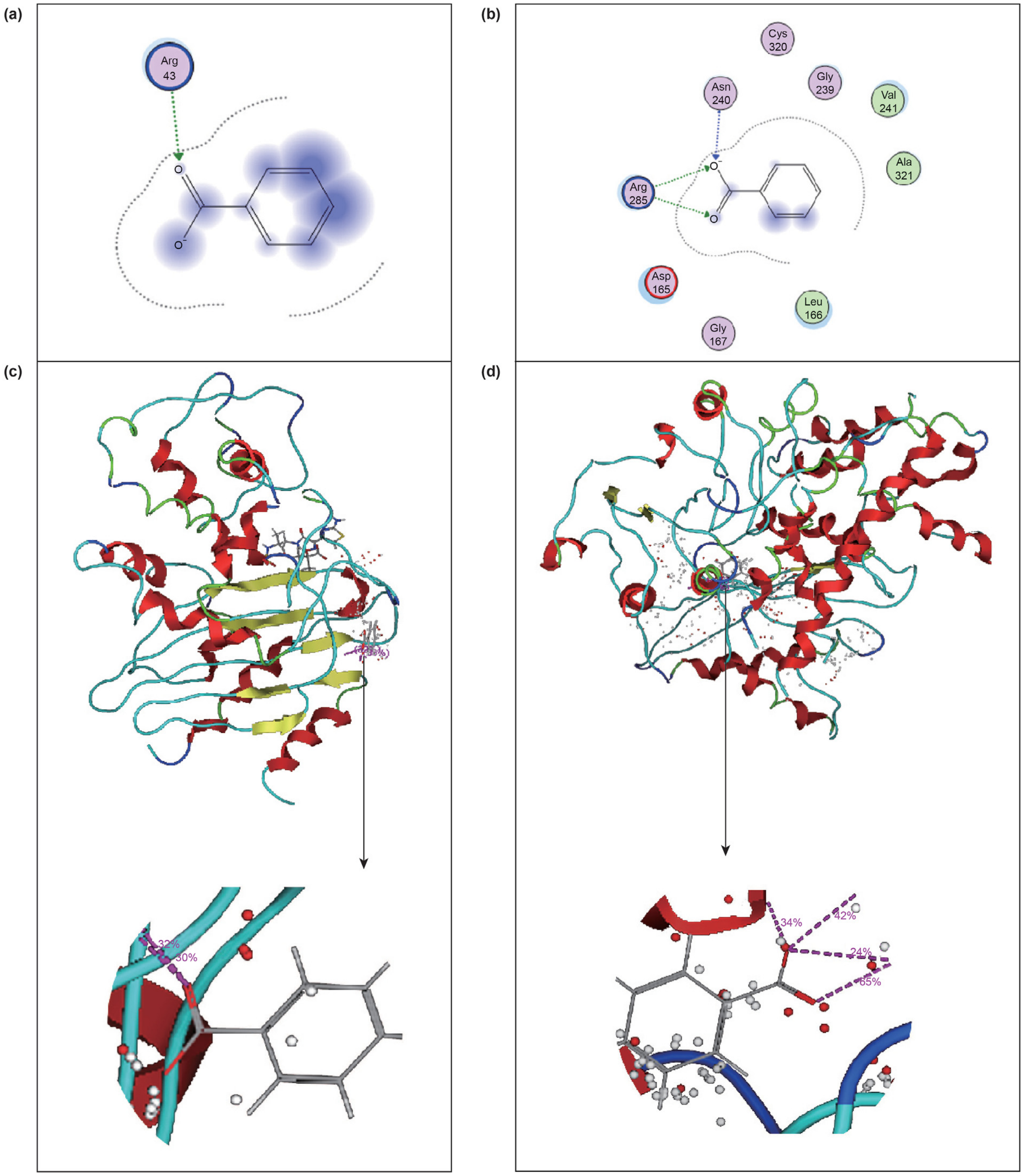

Fig. 5. Molecular simulation of planar diagram (a, b) and space graph (c, d).

acid were connected by two hydrogen bond, and the bond distances are 2.66 Å (32%) and 2.41 Å (30%), respectively. Dehydrogenase and benzoic acid were connected by four hydrogen bond, and the bond distances are 2.51 (34%), 2.67 (35%), 2.86 (42%) and 2.90 (24%), respectively. The immobilised carrier was tightly bound to diesel through hydrogen bonding, and then the diesel was biodegraded by the bacteria after the diesel and dehydrogenase produced by bacteria were combined by hydrogen bonds.

3.3. Adsorption of diesel by Pseudomonas sp. YT-11 strain immobilised on cinnamon shell

In this study, the adsorption of diesel by immobilised petroleum-degradation bacteria was studied. The tight combination of the immobilised carrier, diesel and petroleum-degradation bacteria made the performance of the immobilised petroleum-degradation bacteria superior. The immobilised system was a porous structure and provided an attachment site for petroleum-degradation bacteria. The petroleum-degradation bacteria were immobilised in the carrier to form immobilised petroleum-degradation bacteria. The diesel removal by immobilised petroleum-degradation bacteria depended on adsorption in the initial stage. Diesel was adsorbed in the immobilised carrier surface by hydrogen bonding, thereby promoting the diesel removal from seawater. The adsorption was a spontaneous process. In the immobilised petroleum-degradation bacteria surface, the diesel was absorbed into the petroleum-degradation bacteria by hydrogen bonding and then was biodegraded by the bacteria.

4. Conclusions

In this study, the bacteria immobilised in cinnamon shell were utilised to treat petroleum hydrocarbon pollution. The adsorption of diesel by immobilised petroleum-degradation bacteria was systematically analysed. The results are as follows.

(1) The immobilised petroleum-degradation bacteria were porous materials, and a large number of petroleum-degradation bacteria were attached.
(2) The diesel adsorption conformed to Langmuir adsorption isotherm, which was a spontaneous, entropy-increasing and endothermic process.
(3) The immobilised carrier was tightly bound to diesel through hydrogen bonding. The diesel was biodegraded by the bacteria after the diesel and dehydrogenase produced by bacteria were combined by hydrogen bonds.

Declaration of competing interest

The authors declare that they have no conflict of interest.

Acknowledgments

This work was supported by the scientific research fund project of the National Natural Science Foundation of China (grant numbers 52070123 and 51874193); the Natural Science Foundation of Shandong Province (grant numbers ZR2020ME224); the National Key Research and Development Project (grant number 2019YFC0408503); the Science and Technology Projects of Qingdao (grant number 19-6-2-25-cg); Young Science and Technology Innovation Program of Shandong Province (grant number 2020KJD001); the Open Research Fund Program of Shandong Key Laboratory of Eco-Environmental Science for Yellow River Delta (Binzhou University) (grant number 2019KFJJ02); and Project of Shandong Province Higher Educational Young Innovative Talent Introduction and Cultivation Team [Hydrogen energy chemistry innovation team].

References

Alabresm, A., Chen, Y.P., Decho, A.W., Lead, J., 2018. A novel method for the synergistic remediation of oil-water mixtures using nanoparticles and oil-degrading bacteria. Sci. Total Environ. 630, 1292–1297. https://doi.org/10.1016/j.scitotenv.2018.02.277.

Chandran, P., Das, N., 2011. Degradation of diesel oil by immobilized Candida tropicalis and biofilm formed on gravels. Biodegradation 22, 1181–1189. https://doi.org/10.1007/s10532-011-9473-1.

Chen, Y., Yu, B., Lin, J.J., Naidu, R., Chen, Z.L., 2016. Simultaneous adsorption and biodegradation (SAB) of diesel oil using immobilized Acinetobacter venetianus on porous material. Chem. Eng. J. 289, 463–470. https://doi.org/10.1016/j.cej.2016.01.010.

Du, W.D., Wan, Y.Y., Zhong, N.N., Fei, J.J., Zhang, Z.H., Chen, L.J., Hao, J.M., 2011. Status quo of soil petroleum contamination and evolution of bioremediation. Petrol. Sci. 8, 502–514. https://doi.org/10.1007/s12182-011-0168-3.

Fu, X.G., Wang, H.J., Bai, Y., Xue, J.L., Gao, Y., Hu, S.G., Wu, T.T., Sun, J.K., 2020. Systematic degradation mechanism and pathways analysis of the immobilized bacteria: permeability and biodegradation, kinetic and molecular simulation. Environmental Science and Ecotechnology 2, 100028. https://doi.org/10.1016/j.ese.2020.100028.

Fu, X.G., Wu, T.T., Li, H.S., Xue, J.L., Sun, J.K., Li, L., Qiao, Y.L., Li, C., 2021. Study on the preparation conditions and degradation performance of an efficient immobilized microbial agent for marine oil pollution. Environ. Technol. https://doi.org/10.1080/09593330.2021.1877362.

Fu, X.G., Zhang, Q., Gao, Y., Wu, Y.N., Xiao, X.F., Li, L., Xue, J.L., Liu, B., 2019. Degradation potential of petroleum hydrocarbon-degrading bacteria immobilized on different carriers in marine environment. Petrol. Sci. Technol. 37, 1417–1424. https://doi.org/10.1080/10916466.2019.1587465.

Garcia, A.C.F.S., Araujo, B.R., Birolli, W.G., Marques, C.G., Diniz Jr., L.E.C., Barbosa, A.M., porto, A.L.M., Cruz Romao, L.P., 2019. Fluoranthene biodegradation by Serratia sp. AC-11 immobilized into Chitosan beads. Appl. Biochem. Biotechnol. 188, 1168–1184. https://doi.org/10.1007/s12010-019-02980-9.

Geng, X.L., Boufadel, M.C., Personna, Y.R., Lee, K., Tsao, D., Demicco, E.D., 2014. BIOB: a mathematical model for the biodegradation of low solubility hydrocarbons. Mar. Pollut. Bull. 83, 138–147. https://doi.org/10.1016/j.marpolbul.2014.04.007.

Geng, X.L., Boufadel, M.C., Wrenn, B., 2013. Mathematical modeling of the biodegradation of residual hydrocarbon in a variably-saturated sand column. Biodegradation 24, 153–163. https://doi.org/10.1007/s10532-012-9566-5.

Head, I.M., Jones, D.M., Röling, W.F.M., 2006. Marine microorganisms make a meal of oil. Nat. Rev. Microbiol. 4, 173–182. https://doi.org/10.1038/nrmicro1348.

Helle, I., Ahtiainen, H., Luoma, E., Hanninen, M., Kuikka, S., 2015. A probabilistic approach for a cost-benefit analysis of oil spill management under uncertainty: a Bayesian network model for the Gulf of Finland. J. Environ. Manag. 158, 122–132. https://doi.org/10.1016/j.jenvman.2015.04.042.

Hu, J., Yang, Q., 2015. Microbial degradation of di-n-butyl phthalate by Micrococcus sp immobilized with polyvinyl alcohol. Desalin Water Treat 56, 2457–2463. https://doi.org/10.1080/19443994.2014.963151.

Kim, B.J., Park, S.J., 2007. Influence of surface treatments on micropore structure and hydrogen adsorption behavior of nanoporous carbons. J. Colloid Interface Sci. 311, 619–621. https://doi.org/10.1016/j.jcis.2007.03.049.

Kumar, A.G., Rajan, N.N., Kirubagaran, R., Dharani, G., 2019. Biodegradation of crude oil using self-immobilized hydrocarbonoclastic deep sea bacterial consortium. Mar. Pollut. Bull. 146, 741–750. https://doi.org/10.1016/j.marpolbul.2019.07.006.

Li, N.N., Xu, H.C., Yang, Y.P., Xu, X.M., Xue, J.L., 2019. Preparation, optimization and reusability of immobilized petroleum-degrading bacteria. Environ. Technol. https://doi.org/10.1080/09593330.2019.1703826.

Li, Q.Y., Lu, H., Yin, Y.X., Qin, Y.M., Tang, A.X., Liu, H.B., Liu, Y.Y., 2019. Synergic effect of adsorption and biodegradation enhance cyanide removal by immobilized Alcaligenes sp. strain DN25. J. Hazard Mater. 364, 367–375. https://doi.org/10.1016/j.jhazmat.2018.10.007.

Li, Y.G., Li, W.L., Gao, H.S., Xing, J.M., Liu, H.Z., 2011. Integration of flocculation and adsorptive immobilization of Pseudomonas delafieldii R-8 for diesel oil biodesulfurization. J Chem Techno Biot 86, 246–250. https://doi.org/10.1002/jctb.2510.

Li, Y., Xiong, Y.Q., Yang, W.Y., Xie, Y.L., Li, S.Y., Sun, Y.G., 2009. Compound-specific stable carbon isotopic composition of petroleum hydrocarbons as a tool for tracing the source of oil spills. Mar. Pollut. Bull. 58, 114–117. https://doi.org/10.1016/j.marpolbul.2008.08.012.

Liang, F.Y., Lu, M.M., Keener, T.C., Liu, Z.F., Khang, S.J., 2005. The organic composition of diesel particulate matter, diesel fuel and engine oil of a non-road diesel generator. J. Environ. Monit. : JEM 7, 983–988. https://doi.org/10.1039/b504728e.

Liu, S.L., Wang, Y., Liang, Y.T., 2020. Environmental consequence analysis of oil spills from onshore pipelines with parametric uncertainty. Process Saf Environ 141, 123–134. https://doi.org/10.1016/j.psep.2020.04.032.

Lou, L.P., Huang, Q., Lou, Y.L., Lu, J.R., Hu, B.L., Lin, Q., 2019. Adsorption and degradation in the removal of nonylphenol from water by cells immobilized on biochar. Chemosphere 228, 676–684. https://doi.org/10.1016/j.chemosphere.2019.04.151.

Marin, J.A., Hernandez, T., Garcia, C., 2005. Bioremediation of oil refinery sludge by landfarming in semiarid conditions: influence on soil microbial activity. Environ. Res. 98, 185–195. https://doi.org/10.1016/j.envres.2004.06.005.

Niu, J.F., Xu, J.J., Dai, Y.R., Xu, J.L., Guo, H.Y., Sun, K., Liu, R.L., 2013. Immobilization of horseradish peroxidase by electrospun fibrous membranes for adsorption and degradation of pentachlorophenol in water. J. Hazard Mater. 246, 119–125. https://doi.org/10.1016/j.jhazmat.2012.12.023.

Onwosi, C.O., Odibo, F.J.C., Enebechi, C.K., Nwankwegu, A.S., Ikele, A.I., Okeh, O.C., 2017. Bioremediation of diesel-contaminated soil by Composting with locally generated bulking agents. Soil Sediment Contam. 26, 438–456. https://doi.org/10.1080/15320383.2017.1348337.

Partovinia, A., Rasekh, B., 2018. Review of the immobilized microbial cell systems for bioremediation of petroleum hydrocarbons polluted environments. Crit. Rev. Environ. Sci. Technol. 48, 1–38. https://doi.org/10.1080/10643389.2018.1439652.

Pi, Y.R., Chen, B., Bao, M.T., Zhang, B.Y., 2019. Microbial degradation of four dispersed crude oils by Rhodococcus sp. evaluated using carbon stable isotope analysis. J Chem Technol Biot 94, 1800–1807. https://doi.org/10.1002/jctb.5945.

Shi, K., Zhang, Q., Xue, J.L., Chen, X., Chen, Y.Q., Qiao, Y.L., Yang, Y.P., Sun, J.K., 2020. Study on the degradation performance and bacterial community of bioaugmentation in petroleum-pollution seawater. J Environ Chem Eng 8, 103900. https://doi.org/10.1016/j.jece.2020.103900.

Shi, L., Zhang, D., Zhao, J.F., Xue, J.L., Yin, M.N., Liang, A.P., Pan, B., 2021. New insights into the different adsorption kinetics of gallic acid and tannic acid on minerals via H-1 NMR relaxation of bound water. Sci. Total Environ. 767, 144447. https://doi.org/10.1016/j.scitotenv.2020.144447.

Tang, L.P., Huang, Z.M., Zhu, X.H., Zhou, Y.L., Li, B., 2019. Investigation of the mechanical response of a deep-water drilling riser to ocean currents and waves. Adv. Mech. Eng. 11. https://doi.org/10.1177/1687814018818334.

Wang, X., Wang, X.J., Liu, M., Bu, Y.J., Zhang, J., Chen, J., Zhao, J.F., 2015. Adsorption–synergic biodegradation of diesel oil in synthetic seawater by acclimated strains immobilized on multifunctional materials. Mar. Pollut. Bull. 92, 195–200. https://doi.org/10.1016/j.marpolbul.2014.12.033.

Wu, Y.N., Xu, M., Xue, J.L., Shi, K., Gu, M., 2019. Characterization and enhanced degradation potentials of biosurfactant-producing bacteria isolated from a marine environment. ACS Omega 4, 1645–1651. https://doi.org/10.1021/acsomega.8b02653.

Xiao, X.F., Deng, Y.Y., Xue, J.L., Gao, Y., 2019. Adsorption of chromium by functionalized metal organic frameworks from aqueous solution. Environ. Technol. https://doi.org/10.1080/09593330.2019.1683618.

Xue, J.L., Wu, Y.N., Liu, Z.X., Li, M.L., Sun, X.Y., Wang, H.J., Liu, B., 2017. Characteristic assessment of diesel-degrading bacteria immobilized on natural organic carriers in marine environment: the degradation activity and nutrient. Sci. Rep. 7, 8635. https://doi.org/10.1038/s41598-017-08832-y.

Xue, J.L., Wu, Y.N., Shi, K., Xiao, X.F., Gao, Y., Li, L., Qiao, Y.L., 2019. Study on the degradation performance and kinetics of immobilized cells in straw-alginate beads in marine environment. Bioresour. Technol. 280, 88–94. https://doi.org/10.1016/j.biortech.2019.02.019.

Zahed, M.A., Aziz, H.A., Isa, M.H., Mohajeri, L., Mohajeri, S., Kutty, S.R.M., 2011. Kinetic modeling and half life study on bioremediation of crude oil dispersed by Corexit 9500. J. Hazard Mater. 185, 1027–1031. https://doi.org/10.1016/j.jhazmat.2010.10.009.

Zhang, Y.R., Gao, W., Lin, F.X., Han, B., He, C.F., Li, Q., Gao, X.X., Cui, Z.S., Sun, C.J., Zheng, L., 2018. Study on immobilization of marine oil-degrading bacteria by carrier of algae materials. World J. Microbiol. Biotechnol. 34, 70. https://doi.org/10.1007/s11274-018-2438-3.

Zhou, G.Z., Wang, Y., Zhou, R.S., Wang, C.Z., Jin, Y.Q., Qiu, J., Hua, C.Y., Cao, Y.Y., 2019. Synthesis of amino-functionalized bentonite/$CoFe_2O_4$@MnO_2 magnetic recoverable nanoparticles for aqueous Cd^{2+} removal. Sci. Total Environ. 2682, 505–513. https://doi.org/10.1016/j.scitotenv.2019.05.218.