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Study on wetting mechanism of nonionic silicone surfactant on coal dust

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ABSTRACT

Coal dust disasters are serious in coal mining. The use of nonionic surfactants can effectively improve the wettability of coal dust and reduce the content of suspended coal dust in the air. For the problem of low wettability of ordinary surfactants, this paper selects silicone surfactants with high surface activity and low surface tension to improve the wetting ability of coal dust. To explore the wettability of nonionic silicone surfactants on coal dust, the effects of six nonionic silicone surfactants on the wettability of coal dust surfaces were studied by experiments. The test objects were four kinds of coal samples with different metamorphic degrees. The surface tension, wetting time, and contact angle experiments were carried out, and the critical micelle concentration and the expansion coefficient of the coal surface were calculated. The wetting time of the compound solution was measured to verify the synergistic effect of the compound solution. The results show that: 6 # has the best wetting effect on coal dust, followed by 4 # and 2 #; The order of surface tension is: 1 # < 3 # < 4 # < 6 # < 5 # < 2 #, the surface tension of 1 # is the lowest (19.962 mN/m); 1 # and 4 # are easier to spread on the surface of coal dust, the spreading coefficient of coking coal is the largest and the contact angle is the smallest, which is 18.8°. The 4 # and 6 # with a mass ratio of 8:2 were compounded. The compound surfactant solution had a significant synergistic effect. Compared with the monomer surfactant solution, the wettability of long-flame coal and coking coal increased by 15.14% and 10.00%, respectively. The results of this study can provide reference and experimental support for the development of high-efficiency dust suppressants based on silicone surfactants.

1. Introduction

China is the world's largest producer and consumer of coal. With the continuous increase of coal mining intensity, the dust concentration of underground coal mining face increases sharply. Coal dust has become an important factor threatening underground safety production [1-3]. The existence of coal dust has two hazards: (1) excessive accumulation of coal spontaneous combustion and coal dust explosion; (2) Long-term inhalation can cause severe pneumoconiosis [4-7]. Pneumoconiosis seriously threatens the life and

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health of miners and is irreversible [8]. As of the end of 2021, a total of 915,000 cases of pneumoconiosis have been reported in China, accounting for about 77.65% of the total number of reported occupational diseases. Pneumoconiosis remains the most serious occupational disease among Chinese miners [9,10]. Therefore, the use of effective dust control technology to improve the safety of coal production and maintenance of the health of workers is of great significance [11]. At present, the commonly used dust prevention and control technologies include chemical dust removal, foam dust reduction, spray dust removal, etc. The advantages of high efficiency, convenient construction, and low economic cost make chemical dust removal a current research hotspot.

In chemical dust removal, the addition of surfactant can enhance the wettability of coal dust, effectively reduce the concentration of coal dust, and improve the effectiveness of dust suppression [12,13]. The wettability of coal dust mainly depends on the type of surfactant, the nonionic type has the best dust reduction effect on coal dust, followed by the anion type, and the worst is the cationic type [14]. Therefore, nonionic type surfactants are widely used because of their stronger wettability and better adhesion [15]. To better improve the wettability of coal dust, the researchers compounded the surfactants. Studies have shown that the dust suppression rate of the compound solution is much greater than that of the aqueous solution., which could significantly improve the wettability of coal dust [16]. However, after the common surfactant acts on coal dust, wastewater treatment costs are high and easy to cause pollution to the environment [17]. At present, a large number of researchers have gradually turned their research objectives to dust suppression materials. These dust suppression materials are non-toxic, harmless, and have no secondary pollution.

Silicone surfactants, especially trisiloxane surfactants, because of their high wettability and low surface tension, have attracted the attention of scholars in recent years, The total coverage area of water droplets containing trisiloxane silicone surfactants is very high, which is 25 times that of traditional surfactants [18]. The trisiloxane silicone surfactant not only simple structure, low physiological toxicity, and high surface activity, but also surface tension can be as low as 20 mN/m [19]. Polyether-modified siloxane surfactants can significantly reduce the surface tension of the water system or the surface tension of the non-aqueous system. In the oil system, the surface tension of the solution can be reduced to below 25 mN/m, which greatly improves the oil removal rate [20]. Silicone surfactants have significant advantages in reducing surface tension: strong wettability, high permeability, safe degradability, and high surface activity [21]. But there are few studies on the application of coal dust suppressants [22]. Therefore, it is of practical significance to study silicone surfactants to improve the wettability of underground coal dust.

The purpose of this paper is to explore the wettability of non-ionic silicone surfactants on coal dust. Taking four kinds of coal samples with different metamorphic degrees as test objects, the effects of six kinds of nonionic silicone surfactants on the wettability of coal dust surface were studied experimentally. Using the data of surface tension, the wettability of the monomer solution can be preliminarily judged. The wetting time and static contact angle data are used to calculate the critical surface tension and spreading coefficient. Based on this, the influence of solution concentration on the wettability of coal dust is studied. In addition, the monomer solution with better wettability was compounded, and the wetting time of the compound solution and the monomer solution was compared to explore the synergistic effect of the surfactant. The research results can improve the wettability of coal dust, and provide a reference for the application of nonionic silicone surfactants in coal dust suppressants.

2. Experimental system and test method

2.1. Selection of experimental coal samples

To study the general wettability of nonionic silicone surfactants on coal dust. In this experiment, we selected four different metamorphic coal samples of gas coal, fat coal, long flame coal, and coking coal, and removed some impurities in the coal samples. We crushed four kinds of coal samples with a pulverizer for 3 min and then sieved them into 200 mesh coal dust by industrial sieve, dried them in a vacuum oven at 50 °C for 48 h, and placed them in a sealed bag for storage. The industrial analysis of the four coal samples is shown in Table 1.

2.2. Experimental system and process

Because nonionic surfactants have the best wetting effect compared with anionic and cationic surfactants, and silicone surfactants have the characteristics of high wettability [23–25], Therefore, six nonionic silicone surfactants were selected in this experiment to verify their high wettability and low surface tension to coal dust. Silicone surfactants used in the experiment are shown in Table 2.

In this paper, we will prepare different concentrations of the solution, measure the surface tension of the solution, calculate the critical micelle concentration and critical surface tension of the solution, and verify the low surface tension characteristics of the silicone surfactant solution; We designed the sedimentation experiment and contact angle measurement experiment. We measured the wetting time and contact angle parameters of different concentration solutions, calculated the spreading coefficient of the solution on

Table 1	
Industrial analysis of experimental coal samples.	

serial number	coal samples	M _{ad} (%)	A _{ad} (%)	V_{daf} (%)	FC _d (%)
1	gas coal	1.31	20.44	38.24	48.97
2	fat coal	1.01	20.53	33.48	52.72
3	long-flame coal	8.82	4.60	35.59	55.77
4	coking coal	13.35	10.00	34.05	50.55

Table 2
Six silicone surfactants selected in this experiment.

serial number	name
1#	polyether-modified polysiloxane
2#	Polyether Modified Polydimethylsiloxane
3#	Polyether ester-modified trisiloxane
4#	polyether modified trisiloxane
5#	polydimethylsiloxane
6#	Polyalkylene oxide modified heptamethyltrisiloxane

the coal surface, and compared and verified the super wettability of the silicone surfactant solution; Surfactant compounding can improve the wettability of coal dust. Therefore, two surfactants with the best wettability are selected to compound at a certain ratio. We measure the wetting time of the compound solution and compare the best compounding scheme. The experimental flow chart is shown in Fig. 1. To meet the actual situation of the site, the solution water used in this study is tap water.

2.3. Surface tension determination

Surface tension is an important parameter affecting the dust suppression effect. The lower the surface tension is, the better the wettability of coal dust is [26,27]. In this experiment, we use JYW-200B automatic interfacial tension meter to measure the surface tension of surfactant solution at different concentrations. A total of six concentrations, namely 0.05%, 0.1%, 0.15%, 0.3%, 0.5%, and 0.7%, were set up to measure the surface tension of 24 groups. To ensure the accuracy of the data, each group of experiments was repeated three times to obtain the average surface tension value. After measuring the surface tension data, the critical micelle concentration and critical surface tension of the six solutions were calculated, and then the two were compared. To reduce the experimental error, the solution temperature of the surface tension measurement experiment is 20 °C.

2.4. Wettability determination

The sedimentation experiment can effectively evaluate the wettability of surfactant on coal dust. The shorter the wetting time of coal dust in surfactant solution, the better the wettability of the solution [28,29]. In this experiment, seven kinds of concentration solutions of 0.05%, 0.1%, 0.15%, 0.2%, 0.3%, 0.4%, and 0.5% were prepared. After the foam on the surface of the solution was exhausted, we used an electronic balance to weigh 100 mL surfactant solution into a dry beaker and then weighed 3 g/200 mesh pulverized coal with weighing paper, poured it into the surface of 100 mL solution, and recorded the time required for pulverized coal



Fig. 1. Flow chart of the experiment.

from contact with the solution to complete immersion, that is, the wetting time of coal dust. The settlement experiment steps of the four coal samples are the same as above. A total of 28 groups of experiments were set up. The experimental temperature was 20 °C, and 100 mL of the aqueous solution was taken as the control group. To ensure the accuracy of the experimental data, each experiment was repeated three times to obtain the average wetting time of coal dust.

2.5. Contact angle measurement

The contact angle is an important parameter to indicate the wettability of a solid surface, which can reflect the interaction between liquid and solid. The smaller the contact angle is, the better the wettability of coal dust is [30]. Based on the solution concentration corresponding to the optimal wetting time of the four coal samples, the contact angle data were measured by an SL200 B standard contact angle meter, and 28 groups of experiments were set up. Taking 200 mesh coal dust of gas coal, fat coal, long flame coal, and coking coal, the contact angle of the solution was tested by instrument. According to the surface tension and contact angle data of the solution, we calculate the spreading coefficients of six solutions on different coal surfaces and compare and verify which surfactant solution has the best wettability to coal dust. At the same time, we measured the contact angle of four kinds of coal dust in an aqueous solution and compared the contact angle of the aqueous solution with the contact angle of the surfactant solution.

2.6. Wettability determination of surfactant compounding

Compared with the monomer solution, the surfactant compound solution can improve the surface activity of the solution, and greatly reduce the critical micelle concentration, and surface tension of the compound system [31]. Considering the factors such as economy and simplicity, we chose two kinds of silicone surfactants with the best wettability, which were compounded at 9 ratios of 1:9, 2:8, 3:7, 4:6, 5:5, 6:4, 7:3, 8:2, and 9:1. We measured the wetting time of four coal samples: gas coal, fat coal, long flame coal, and coking coal. We set up a total of 36 groups of the compound solution sedimentation experiment, the experimental temperature is 20 °C, and to ensure the accuracy of the data, each group of experiments repeated three times to get the average wetting time. By comparing with the wetting time of the monomer solution, the shortest wetting time is obtained, to select the best proportion of the compound scheme.

3. Results and discussion

3.1. Surface tension analysis of solution

The relationship between the surface tension of the solution and the concentration is shown in Fig. 2. It can be seen from Fig. 2 that the change of surface tension with surfactant concentration can be divided into three regions: descending region, transition region, and stable region. In the descending region, the surface tension decreases significantly with the increase of surfactant concentration; In the transition region, the surface tension of the six surfactants from small to large: 1# < 3# < 4# < 6# < 5# < 2#, the surface tension of the six surface tension of the six surface tension, the storage tension of 19.962 mN/m; In the stable region, the surface tension remains almost constant with the increase of surfactant concentration.

It can be found from Fig. 2, the value of surface tension decreases first and then remains stable with the increase in concentration. This is because as the concentration increases, the surfactant molecules on the surface of the solution begin to adsorb, and the surface



Fig. 2. Determination of surface tension of six solutions at different concentrations.

tension decreases significantly. With the further increase of solution concentration, the adsorption density of surfactant molecules at the gas-liquid interface gradually reaches saturation, and the lipophilic group self-polymerizes in the solution to form micelles. At this time, the solution concentration reaches the critical micelle concentration, and the surface tension of the solution tends to be stable after saturation. To further obtain the critical micelle concentration and critical surface tension of each surfactant, a comparison between the six surfactants was carried out. We calculated the logarithm of solution concentration (*lgc*) and further compared the relationship between surface tension and the logarithm of solution concentration. The relationship is shown in Fig. 3 γ_{CMC} is the critical surface tension at the critical micelle concentration.

It can be seen from Fig. 3(a, b) that the critical surface tension of the six silicone surfactants is from small to large: 1# < 3# < 4# < 6# < 5# < 2#; the critical micelle concentration from small to large is: 1# = 6# < 2# = 3# = 4# < 5#, which shows that 1# has the strongest ability to reduce surface tension compared to other surfactants, and the concentration required to form micelles is the smallest, and the wetting effect on coal dust is the best. At the same concentration, 1# had the lowest surface tension of 19.962 mN/m, and the corresponding critical micelle concentration was 1 mmol/L. In addition, the critical micelle concentrations of 1# ~ 6# were determined to be 0.1%, 0.15%, 0.15%, 0.3%, and 0.1%, respectively. In summary, the six silicone surfactants are in line with the expected results from the parameter of surface tension.

3.2. Wetting time analysis of coal dust

The wetting time of different surfactant solutions to coal dust is shown in Fig. 4. It can be seen from Fig. 4 that there are significant differences in the wettability of the four coal samples. The wettability of coking coal is the best, while the wettability of the other three coal samples is poor.

From Fig. 4(a~d), it can be found that six kinds of surfactant solutions to improve the wetting ability of coal dust from strong to weak: 6# > 4# > 3# > 1# > 5# > 2#, 6# has the best wetting effect on coal dust, and 2# has the worst wetting effect. With the increase of solution concentration, the wetting time decreases sharply; When the solution concentration exceeds 0.3%, the wetting time remains relatively stable. Through the changing trend of four kinds of coal samples, it can also be found that the same surfactant has certain differences in the wettability of coal samples with different metamorphic degrees. The better the wettability of coal samples, the lower the improvement rate of wettability. The better the wettability of coal samples, the lower the improvement rate of wettability improvement rate. For example, 6# to the best wettability of coal increased by 39.41%, but to the poor wettability of gas coal, fat coal, and flame coal increased by 54.68%, 52.21%, and 49.83% respectively.

It can be seen from Figs. 2–4 that, the decrease in surface tension can reduce the energy barrier when coal dust enters the solution, thus improving the wettability of coal dust. When the solution concentration exceeds the critical micelle concentration, the surface tension remains stable, but the wetting time still shows a downward trend, indicating that the wetting time of coal dust is no longer related to surface tension. The wetting process of coal dust in a nonionic silicone surfactant solution is shown in Fig. 5. At this time, with the increase of solution concentration and the number of hydrophilic groups on the surface of coal dust, the adsorption density of surfactant increases, so the wettability of coal dust is further improved and the wetting time is gradually shortened.

The optimum wetting concentrations of different surfactants on four coal samples are shown in Table 3.



(a) 1#, 2#, 3#



Fig. 3. Variation of surface tension with lgc.



Fig. 4. Wetting time of six surfactant solutions to coal dust.

According to the best wetting concentration of four kinds of coal samples, we compare the best wetting concentration and the initial concentration of wettability under the rate of increase and the best wetting time, screening the best wettability of silicone surfactant. The enhancement rate of surfactant solution on the wettability of coal dust is shown in Fig. 6.

It can be seen from Fig. 6 that, compared with the initial concentration, the improvement rate of wettability of the 2 # solution is the highest, but at the optimal wetting concentration, the 2# solution has the longest wetting time. Among the six surfactant solutions, the 6# solution has the shortest wetting time for gas coal, fat coal, and long flame coal, and the 4# solution has the shortest wetting time for coking coal. However, in comparison, the wetting time of coking coal in 4# solution is 20.98 s, 6# solution is 21.28 s, and the wetting time is only 0.3 s. Therefore, 6# to reduce the wetting time of the four kinds of coal dust best performance.

3.3. Contact angle analysis of coal samples

Based on the optimum wetting concentrations of the four coal samples, we used aqueous solution as the control group and then measured the contact angle at the optimal wetting concentration. The contact angle data are shown in Table 4.

From Tables 4 and it can be seen that the contact angle between the surfactant solution the and coal sample is lower than that between water and coal samples, but the degree of reduction is different. The smaller the contact angle, the better the wettability of the solution. The experimental results are shown in Fig. 7. When $C_{1\#}$ is 0.3%, the minimum contact angles of fat coal and long flame coal



Fig. 5. Changes in the adsorption state of surfactant molecules with solution concentration.

 Table 3

 Optimum wetting concentrations of surfactants on four coal samples (%).

coal samples	1#	2#	3#	4#	5#	6#
gas coal	0.4	0.4	0.4	0.3	0.3	0.4
fat coal	0.3	0.4	0.3	0.3	0.3	0.3
long-flame coal	0.3	0.3	0.3	0.3	0.3	0.3
coking coal	0.4	0.5	0.5	0.4	0.5	0.4



Fig. 6. The improvement rate of wettability of coal dust under the optimum wetting concentration.

are 32.9° and 31.2° respectively. When $C_{4\#}$ is 0.3% and 0.4% respectively, the minimum contact angles of gas coal and coking coal are 29.7° and 18.8° respectively. After the action of surfactants, the contact angles of gas coal, fat coal, long-flame coal, and coking coal decreased by 59.09%, 61.61%, 48.60%, and 71.12%, respectively. This indicates that the wettability of coal dust is greatly improved after surfactant acts on the coal surface. Among them, 1# and 4# have the best effect on the contact angle of coal dust.

It can be seen from Figs. 2–7 that, according to the data of surface tension, wetting time, and contact angle, the silicone surfactants with the best wetting effect under each parameter are not completely consistent. This is because the determination of surface tension is in the environment of air and liquid, and the determination of wetting time and contact angle is in the coexistence of air, coal sample, and liquid. Therefore, wetting time and contact angle need to consider the interaction the between coal sample and surfactant.

3.4. Analysis of the spreading coefficient of the solution

According to the surface tension and contact angle data measured in this paper, the spreading coefficients of $1\# \sim 6\#$ surfactants on four coal samples were analyzed, as shown in Fig. 8. The larger the spreading coefficient, the better the wettability of the solution.

Table 4

Contact angles of four coal samples at an optimum wetting concentration (°).

concentration	water	1# (0.4%)	2# (0.4%)	3# (0.4%)	4# (0.3%)	5# (0.3%)	6# (0.4%)
gas coal	72.6	44.5	41.9	35.4	29.7	32.1	34.3
concentration	water	1#(0.3%)	2#	3# (0.3%)	4#	5# (0.3%)	6# (0.3%)
			(0.4%)		(0.3%)		
fat coal	85.7	32.9	38.0	48.8	35.6	38.6	36.7
concentration	water	1#(0.3%)	2#	3# (0.3%)	4#	5# (0.3%)	6# (0.3%)
			(0.3%)		(0.3%)		
long-flame coal	60.7	31.2	41.9	42.2	40.5	39.3	41.8
concentration	water	1#(0.4%)	2#	3# (0.5%)	4#	5# (0.5%)	6# (0.4%)
			(0.5%)		(0.4%)		
coking coal	65.1	36.7	37.6	36.5	18.8	30.8	20.9



Fig. 7. Determination of contact angle of four coal samples.

When $S \ge 0$, it indicates that the solution can spread and wet spontaneously; Conversely, it indicates that the solution cannot spontaneously spread and wet. The calculation formula of the spreading coefficient is as follows:

$$S = \gamma_{la}(\cos \theta - 1) \#$$

where S is the spreading coefficient, mN/m; γ_{la} is the surface tension of surfactant solution, mN/m; θ is the contact angle, °.

It can be seen from Fig. 8 that, the spreading coefficients of the six silicone surfactants are all less than 0, indicating that the six surfactant solutions cannot spontaneously diffuse on the coal surface. When $C_{1\#} = 0.3\%$, the spreading coefficient of fat coal and long flame coal is the largest; When $C_{4\#}$ is 0.3% and 0.4% respectively, the spreading coefficient of gas coal and coking coal is the largest.



Fig. 8. Relationship between spreading coefficient and surfactant.

The results show that 1# is easier to spread on the surface of fat coal and long flame coal; 4# is easier to spread on the surface of gas coal and coking coal. It can also be seen from Fig. 8 that, among the four coal samples, the spreading coefficient of coking coal with the best wettability is-1.07, and the spreading coefficient is the largest among the four, which is consistent with the contact angle measurement data.

3.5. Wetting time analysis of surfactant mixture

According to the measurement data of the wetting time of surfactant monomer, we studied the compounding of surfactants. It can be seen from Fig. 4 that the wetting effect of 4 # and 6 # on four coal samples is the best. Therefore, we compounded with the best wetting concentration at 9 ratios, the total proportion of surfactants was 0.3%, and the proportion of water was 99.7%. We measured the wetting time of coal dust in the compound solution. The experimental data of the compound surfactant solution and the monomer surfactant are shown in Fig. 9.

It can be seen from Fig. 9(a, b) that the compound solution has no promoting effect on the wettability of gas coal and fat coal. However, it can be found from Fig. 9 (c, d) that the wetting time of the compound solution is mostly lower than that of the monomer solution for long flame coal and coking coal, which indicates that the combination of surfactants has a synergistic effect. When the ratio of 4# and 6# is 8:2, the wetting time of long flame coal and coking coal are the lowest, and the wettability is increased by 15.14% and 10.00% respectively compared with monomer solution. Through compounding experiments, it can be found that the compound solution is not necessarily better than the wettability of the monomer, because this is determined by the nature of the surfactant and the coal sample itself.

3.6. Comparative discussion analysis

In terms of surface tension, Wang et al. measured the surface tension of SDBS ordinary surfactant at 0.5% concentration was 28.0 mN/m, and its critical micelle concentration was about 0.005% [32]. Zhou et al. selected surfactants to be compounded with magnetized water, and the lowest surface tension was measured to be 28.070 mN/m [28]. Wang et al. selected rhamnolipid and lactone sophorolipid biosurfactants. When the compounding ratio was 9:1, the minimum surface tension was 28.515 mN/m [17]. Jiang et al. measured that SDS had a higher efficiency in reducing surface tension through surface tension experiments. When the surface tension was 0.5% [33]. However, this paper selects 1 # solution with 0.1% concentration, and the minimum surface tension is 19.962 mN/m. The surface tension pair is shown in Fig. 10. Compared with previous studies, the surface tension of the silicone surfactant studied in this experiment is the lowest, and the solution concentration is also low, which achieves the expected research purpose.

From the results, we selected and compounded the obtained silicone surfactant solution in this paper. We found that this solution is superior to the surfactant used in general production in terms of surface tension, which can effectively reduce the mine dust concentration and ensure the health and safety of underground workers. However, silicone surfactants are rarely used in coal mine production. At present, the safety, degradability, mild non-toxicity, and environmental friendliness of silicone surfactants need to be further studied.





4. Conclusion

- (1) Based on the surface tension, coal dust deposition, and contact angle experiments, the data of surface tension, coal dust wetting time, and contact angle were analyzed. We studied the effect of a nonionic silicone surfactant aqueous solution system on coal dust wetting. When the critical micelle concentration is 1 mmol/L, the surface tension is as low as 19.962 mN/m, which promotes the adsorption of water molecules on the coal surface.
- (2) The results of the sedimentation experiment show that 4 # and 6 # have the best effect on improving the wettability of coal dust; The contact angle experiment shows that after adding 1 # and 4 # surfactants to the aqueous solution, the contact angles of gas coal, fat coal, long flame coal, and coking coal are reduced by 59.09%, 61.61%, 48.60%, and 71.12%, respectively, which increases the interaction between the coal sample surface and water, and the analysis of the spreading coefficient is consistent with the results of the contact angle.
- (3) We used the method of comparing the surfactant monomer and the surfactant compound and proved that the compound solution had a synergistic effect. When the mass concentration was 0.24% of 4 # and 0.06% of 6 #, the wettability of long flame coal increased by 15.14%, coking coal increased by 10.00%, the wetting time decreased and the wetting effect was higher than that of monomer silicone surfactant, which effectively improved the wettability of coal dust.



Fig. 10. Comparison of surface tension of different surfactants.

Author contribution statement

Kai Wang: Conceived and designed the experiments; Analyzed and interpreted the data. Peiyu Jing: Performed the experiments; Contributed reagents, materials, analysis tools or data; Wrote the paper. Hongfei Qu: Analyzed and interpreted the data. Lulu Huang: Performed the experiments. Chaojie Liu; Zhijing Wang: Contributed reagents, materials, analysis tools or data; Wrote the paper.

Data availability statement

Data included in article/supp. material/referenced in article.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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K. Wang et al.

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