Electronic Supplementary Material

Self-cleaning of superhydrophobic nanostructured surfaces at low humidity enhanced by vertical electric field

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1 Supplementary video

Video ESM1. Droplet impact and rebound on hydrophobic surface. A water droplet impacts on a hydrophobic surface. The water contact angle of the substrate is 158°. The process is recorded at 6,400 frames per second (fps) and shown at 200 fps. The movie corresponds to the test in Fig. 2(e).

Video ESM2. Electrohydrodynamic aggregation behaviour of wet pollution. A pollution (Fe₂(SO₄)₃ particles) was scatter and uniform wetting on the super hydrophobic surface. The applied electric field intensity is 5 kV cm⁻¹. The water contact angle of the substrate is 158° and the pollution contact angle of the substrate is 155°. The process time of aggregation behaviour is 14 s. The movies correspond to the tests in Figs. 3(a) and 3(b).

Video ESM3. Electrohydrodynamic jumping behaviour of wet pollution. A pollution (Fe₂(SO₄)₃ particles) was scatter and uniform wetting on the super hydrophobic surface. The applied electric field intensity is 7.3 kV cm⁻¹. The water contact angle of the substrate is 158° and the pollution contact angle of the substrate is 155°. The process time of aggregation behaviour is 320 ms. The movies correspond to the tests in Figs. 4(c) and 4(d).

Video ESM4. Electrohydrodynamic sliding behaviour of wet pollution. A pollution (Fe₂(SO₄)₃ particles) was scatter and uniform wetting on the super hydrophobic surface. The applied electric field intensity is 4 kV cm⁻¹. The slope of substrate is 1.5°. The water contact angle of the substrate is 158° and the pollution contact angle of the substrate is 155°. The process time of aggregation behaviour is 9.1 s. The movies correspond to the tests in Figs. 4(e) and 4(f).

Video ESM5. Practical application of self-cleaning assisted by an electric field. The pollution (Fe₂(SO₄)₃ particles) was scatter and uniform wetting on the super hydrophobic surface. The applied electric field intensity is 4 kV cm⁻¹. The slope of substrate is 1.5°. The water contact angle of the substrate is 158° and the pollution contact angle of the substrate is 155°. The movies correspond to the tests in Figs. 5(b) and 5(c).

2 Supplementary figures

Figure S1 Roughness of the super hydrophobic surface in AFM image. (a) Surface roughness profile. (b) Distribution of roughness.

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Figure S2  Roughness of the silicone rubber substrate surface in AFM image. (a) Surface roughness profile. (b) Distribution of roughness.

Figure S3  Curve of the electric field intensity versus aggregation time. The dots represent the test points at different humidities and the curve is the curve drawn by fitting calculation results at different humidities. The maximum electric field intensity is 30 kV·cm⁻¹ due to air gap breakdown. With increasing humidity, the aggregation time is significantly reduced and the critical electric field intensity conditions are also reduced.

Figure S4  The moisture absorption rate characteristics of Fe₄(SO₄)₃ with different humidities. The dots represent the test points at different humidities. With increasing humidity, the moisture absorption rate of pollution increases gradually, as does the rate of moisture absorption rate. When the humidity is low, the overall moisture absorption rate is unchanged significantly. When the humidity is higher than 6.4 g·kg⁻¹, the moisture absorption rate and its increasing rate clearly increase. The moisture absorption rate in this paper is 0.05 g·g⁻¹, which indicates that the pollution is in a solid-liquid mixed state.
Figure S5  The aggregation characteristics of different pollutants. The area of the pollution is reduced in the wetting state. In the electric field environment, aggregation can be seen. The area under the electric field is significantly lower than that under the initial condition and wetting state. At the same time, diatomite has the largest reduction, and the aggregation characteristics are the most obvious.

Figure S6  The test circuit schematic.
## 3 Supplementary table

| Applications          | Materials                                                                 | Environment                                       | Droplet features                                                                 | Refs |
|-----------------------|---------------------------------------------------------------------------|---------------------------------------------------|-----------------------------------------------------------------------------------|------|
| Anti- icing           | Concrete and polyvinyl alcohol fibers modified with water-based siloxane emulsion | Temperature: -20°C                                | Dews were frozeed and hard to escape                                               | S1   |
|                       |                                                                             | Humidity ratio: 10.9 g kg\(^{-1}\)                | Dripping droplets were hard to adhere                                              |      |
|                       |                                                                             |                                                   |                                                                                    |      |
|                       | Silica nanoparticles                                                       |                                                   |                                                                                    |      |
|                       |                                                                             |                                                   |                                                                                    |      |
| Anti- icing           | Stainless steel and zinc oxide nano particles modified with fluorinated molecules | Temperature: -10°C                                | Dews were frozeed and hard to escape                                               | S2   |
|                       |                                                                             | Humidity ratio: 18.8 g kg\(^{-1}\)                | Dripping droplets were hard to adhere                                              |      |
|                       |                                                                             |                                                   |                                                                                    |      |
| Anti- icing           | Cationic poly (2-(methacryloyloxy)-ethyltrimethylammonium) and anionic poly (3-sulfopropyl methacrylate) brushes used on gold surface, modified with either initiator or methyl-terminated monolayers | Temperature: -25°C                                | Dews were frozeed and hard to escape                                               | S3   |
|                       |                                                                             | Humidity ratio: 22.0 g kg\(^{-1}\)                | Dripping droplets were discretely distributed.                                    |      |
|                       |                                                                             |                                                   | Droplets had ice nucleation                                                        |      |
|                       |                                                                             |                                                   |                                                                                    |      |
| Anti- icing           | Poly(dimethylsiloxane) and zinc oxide nanoparticles modified with fluorinated molecules | Temperature: -20°C                                | Surface was dry and the swollen ice was merely confined at the edge of the surface | S4   |
|                       |                                                                             | Humidity ratio: 0.6 g kg\(^{-1}\)                 | Self-cleaning and droplets was easy to escape                                      |      |
|                       |                                                                             |                                                   |                                                                                    |      |
| Anti- icing           | Flexible super hydrophobic surface based on natural lotus-leaf surface     |                                                   |                                                                                    |      |
|                       |                                                                             |                                                   |                                                                                    |      |
| Anti- icing           | Fluorinated silicon                                                         | Temperature: -15°C and -25°C                       | Surface keep dry in -15°C and wetted in -25°C                                      | S5   |
|                       |                                                                             | Humidity ratio: 0.7 g kg\(^{-1}\)                 | No droplets formation                                                             |      |
|                       |                                                                             |                                                   | a little frost when wetting                                                        |      |
| Anti- fogging and decontamination | Copper and copper oxide micro-nano structured substrate modified with fluorinated molecules | Temperature: 1.7°C                                | Surface formed a large number of discrete droplets                                | S6   |
|                       |                                                                             | Humidity ratio: 36.7 g kg\(^{-1}\)                | Dews were easy to escape                                                           |      |
|                       |                                                                             |                                                   | Surface formed droplets                                                            |      |
| Anti- fogging and decontamination | Titanium dioxide micro-nano structured substrate modified with fluorinated molecules | Temperature: 3°C                                | Dews were easy to escape                                                           | S7   |
|                       |                                                                             | Humidity ratio: 19.1 g kg\(^{-1}\)                | No droplets generated on surface                                                  |      |
| Anti- fogging and decontamination | Nickel and aluminium substrate                                              | Temperature: 25°C                                | Dripping droplets bounce could take away the pollution                            | S8   |
|                       |                                                                             | Humidity ratio: 11.9 g kg\(^{-1}\)                |                                                                                    |      |
|                       |                                                                             |                                                   |                                                                                    |      |
|                       | Commercial spray consisting of nanoparticles                               |                                                   |                                                                                    |      |
|                       |                                                                             |                                                   |                                                                                    |      |
| Applications                      | Materials                                                                 | Environment                                                                 | Droplet features                                                                 | Refs |
|----------------------------------|---------------------------------------------------------------------------|-----------------------------------------------------------------------------|----------------------------------------------------------------------------------|------|
| Anti-fogging and decontamination | Silicon micro-nano structured substrate modified with fluorinated molecules | Temperature: 10°C Humidity ratio: 18.0 g kg⁻¹                             | Surface formed a large number of discrete droplets Droplets were easy to escape  | S9   |
| Anti-fogging and decontamination | Copper micro-nano structured substrate modified with methyltriethoxysilane | Temperature: 35°C Humidity ratio: 34.7 g kg⁻¹                             | No droplets generated on surface Dripping droplets bounce could take away the pollution | S10  |
| Anti-fogging and decontamination | steel micro-nano structured substrate modified with tetraethyl orthosilicate, trimethylethoxysilane and methyltriethoxysilane | Temperature: 30°C Humidity ratio: 30.1 g kg⁻¹                             | Surface had discrete small droplets Pollution was removed needed droplets motion Small droplets was hard to take away the pollution | S11  |
| Salt spray and decontamination   | mild steel substrates Imidazole encapsulation into lumen of hollow zinc phosphate nanoparticles | Temperature: 30°C Humidity ratio: 38.8 g kg⁻¹ (5 wt.% sodium chloride solution) | Surface formed a large number of discrete droplets Pollution was removed needed droplets motion | S12  |
| Salt spray and decontamination   | mild steel substrates Commercial super hydrophobic spray | Temperature: 35°C Humidity ratio: 40.4 g kg⁻¹ (3.5 wt.% sodium chloride solution) | Surface formed discrete droplets Pollution was removed needed droplets motion | S13  |
| Rainfall and decontamination     | Waterborne silicone resin emulsion and silicon dioxide nanoparticles modified with fluorinated molecules | Temperature: 31°C Rainfall: 25.2 mm 24h⁻¹                             | Most of the impacting raindrops bounced off surface Dripping droplets bounce could take away the pollution | S14  |
| decontamination                   | Polyethylene films Stretching-controlled micromolding process | Temperature: 18°C Humidity ratio: 5.1 g kg⁻¹ (Dry processing) | No droplets generated on surface Pollution deposited and did not remove without other effect Pollution was difficult to remove without droplets motion | S15  |
Table S2 The simulation parameters

| The simulation parameters | Value          |
|---------------------------|----------------|
| The modulus of elasticity of pollution $E_i$ | 8 MPa          |
| The Poisson ratio of pollution $\nu$          | 0.25           |
| The elastic deformation energy constant $k_a$ | 3              |
| The vacuum dielectric constant $\varepsilon_0$ | $8.85 \times 10^{-12} F / m$ |
| The relative dielectric constant of water $\varepsilon_{r1}$ | 81             |
| The relative dielectric constant of air $\varepsilon_{r2}$ | 1.006          |
| The relative dielectric constant of ferric sulfate | 14.2           |
| The initial surface tension of pollution   | $7.3 \times 10^{-2} N / m$ |
| Pressure                                | 97 kPa         |
| The temperature                         | 303.15 K       |

where elastic deformation energy constant comes from the book of *physical mechanics of surface and interface*[S16], the modulus of elasticity and Poisson ratio of pollution are obtained by using the strain controlled triaxial system (VJ Tech Ltd., United Kingdom), which can test the powder, the relative dielectric constant of materials come from the dielectric constant table of Honeywell*[S17,S18].

4 Supplementary Note

Note S1: Analysis and calculation of the wet air properties.

The influence of the humidity

Environmental humidity is one of the key factors affecting the application of superhydrophobic materials*[S19]. When performing conventional experiments, many scholars typically use the relative humidity and temperature as the standard environmental conditions. However, the environmental humidity was different in the same relative humidity environment in different experiments. The environmental humidity of different experiments is difficult to directly evaluate by using the relative humidity. The humidity ratio is an important standard parameter to evaluate the environmental humidity and has a certain conversion relationship with the relative humidity. Therefore, in order to uniformly measure the humidity conditions in different test environments, it is necessary to convert the relative humidity into the humidity ratio to evaluate the environmental humidity for analysis and explanation.

The calculation of the humidity ratio

This article used the American Society of Heating Refrigerating and Airconditioning Engineer (ASHRAE) Handbook Fundamentals SI Edition*[S20] to calculate and analyse the humidity ratio.

The humidity ratio $W$ of a given moist air sample is defined as the ratio of the mass of water vapour to the mass of dry air in the sample, as equation (S1).

$$W = \frac{M_{H_2O}}{M_{ar}} \frac{p_w}{p - p_w}$$  \hspace{1cm} (S1)

where $M_{H_2O}$ is the relative molecular mass of water, $M_{ar}$ is the relative molecular mass of air, $p_w$ represents the partial pressure of water vapour, and $p$ represents the total mixture pressure of dry air.

The relative humidity $\xi$ is defined in equation (S2).

$$\xi = 100\% \frac{p_w}{p_{sw}}$$  \hspace{1cm} (S2)

where $p_{sw}$ represents the saturation pressure of water vapour in the absence of air at the given temperature.

Combining equations (S1) and (S2) the following:

$$W = \frac{M_{H_2O}}{M_{ar}} \frac{\phi p_{sw}}{p - \phi p_{sw}}$$  \hspace{1cm} (S3)

The Hyland-Wexler formula is used to calculate the saturation pressure of water vapour. The equation is as follows:

$$\ln p_{sw} = \begin{cases} \frac{C_1}{T} + C_2 + C_3 T + C_4 T^2 + C_5 T^3 + C_6 T^4 + C_7 \ln T & -100^\circ C \leq T \leq 0^\circ C \\ \frac{C_8}{T} + C_9 + C_{10} T + C_{11} T^2 + C_{12} T^3 + C_{13} \ln T & 0^\circ C \leq T \leq 200^\circ C \end{cases}$$  \hspace{1cm} (S4)

The relevant calculation coefficients are shown in Table S3.
Table S3 The calculation coefficients of equation (S4)

|   | C1     | C2     | C3     | C4     | C5     | C6     |
|---|--------|--------|--------|--------|--------|--------|
| 1 | -5.7x10^{-3} | 6.4 | -9.7x10^{-5} | -6.2x10^{-1} | 2.1x10^{-4} | -9.5x10^{-13} | 4.2 |
| 2 | C8     | C9     | C10    | C11    | C12    | C13    |
| 3 | -5.8x10^{-1} | 1.4 | -4.9x10^{-2} | 4.2x10^{-3} | -1.4x10^{-4} | 6.5 |

Combining equations (S3) and (S4), the humidity ratio in the test environment can be calculated by using the ambient temperature and relative humidity. The calculation results are shown in Fig. 5(d) and Table S3. However, due to a large number of scholarly studies, the ambient pressure in the test was not given, and the ASHRAE Handbook calculated the parameters based on the standard air pressure environment (101.325 kPa). For this reason, the calculation results in this paper are all analysed with the standard air pressure, and there are some errors between the calculation results and the actual test environment.

Note S2: Mechanical analysis of droplet aggregation

In order to further research the self-aggregation characteristics of pollution, fluid dynamics were adopted. We mainly used the finite element and phase field methods to study the pollution dynamics effects on the superhydrophobic surface. The theory of electro-elasto-capillarity dynamics was established as follows, and a schematic diagram of the model is shown in Fig. 4(a).

The change energy of the pollution system

The pollution of spontaneous aggregation has a critical surface tension, which can be described as follows[S21]:

\[ L_{EC} = \frac{D}{\sqrt{(1 + \cos \theta)} \gamma_{n}} \]  

(S5)

where \( L_{EC} \) is the critical constant of the water droplet, \( \theta \) is the contact angle of the water droplet and the insulator surface, \( \gamma_{n} \) is the surface tension, and \( D \) is the bending rigidity of pollution, which can be calculated as follows.

\[ D = \frac{E h^{3}}{12(1 - v^{2})} \]  

(S6)

where \( E \) is the modulus of elasticity of pollution, \( v \) is the Poisson ratio, \( h \) is the thickness of pollution.

In the process of pollution aggregation, the interface energy of the water droplet was reduced and converted to elastic deformation energy. When the interface energy was larger than the elastic deformation energy, the pollution could spontaneously aggregate. The energy conversion of the two-dimensional model was calculated as follows[S16].

\[ \Delta E_{interface} = \gamma_{n} (\int l_{d}dl - \int l_{s}dl) = \gamma_{n} (2L + 2h - 2\pi R) \]  

(S7)

\[ \Delta E_{bending} = k_{s} \frac{DL}{R^{2}} \]  

(S8)

\[ \Delta E_{v} = \Delta E_{interface} - \Delta E_{bending} \]  

(S9)

where \( \Delta E_{interface} \) is the change in the interface energy, \( \int l_{d}dl \) is the perimeter of the water droplet at the initial state, \( \int l_{s}dl \) is the perimeter of the pollution droplet at the final state, \( \Delta E_{bending} \) is the change in the elastic deformation energy, \( k_{s} \) is the elastic deformation energy constant, and \( \Delta E_{v} \) is the change in the system energy.

In the process of pollution wetting, the perimeter of the water droplet was less than the critical constant of the water droplet, due to the limitation in the environmental humidity. Therefore, the droplet could not be spontaneously aggregated. However, when the water droplet was in the electric field, the surface tension of the water droplet increased with increasing electric field intensity due to the polarization of the water droplet. According to Bateni’s experimental results, the surface tension of water droplet can be described as follows[S22].

\[ \gamma_{n} = \gamma_{n0} + \frac{\varepsilon_{0} \varepsilon_{r} E^{2}}{4b} \]  

(S10)

where \( \gamma_{n0} \) is the surface tension of the water droplet at its initial state, \( b \) is the surface curvature of the water droplet, \( \varepsilon_{0} \) is the dielectric constant of vacuum, \( \varepsilon_{r} \) is the dielectric constant of pollution, and \( E \) is the electric field intensity.

According to (S5) and (S10), the relationship between the electric field intensity and the critical constant can be obtained.

\[ L_{EC} = \frac{D}{\sqrt{(1 + \cos \theta)(\gamma_{n0} + \frac{\varepsilon_{0} \varepsilon_{r} E^{2}}{4b})}} \]  

(S11)

With the increase of the electric field intensity, the critical constant was reduced. This reduction resulted in the pollution spontaneously aggregating under the condition of fewer water droplets. Furthermore, the relationship between the decrease in the critical constant and the increase in the electric field intensity was directly proportional.

To further research the self-aggregation characteristics of pollution, fluid dynamics was adopted for the two-dimensional simulation analysis. Additionally, the Maxwell stress of the droplet polarization and the model of the energy conversion were considered in an electric field environment. The simulation model is described below (the detailed calculation analysis is provided
in Note S3).

\[ \rho \frac{\partial u}{\partial t} + \rho (u \cdot \nabla) u = \nabla \left\{ -pI + \mu (\nabla u + (\nabla u)^T) \right\} + F_e + \rho g + F_i \tag{S12} \]

\[ F_e = \nabla \cdot T = \nabla \cdot (\varepsilon_r \varepsilon_0 E \cdot E^T - \frac{1}{2} E^2 I) \tag{S13} \]

\[ \frac{\partial \phi}{\partial t} + u \cdot \nabla \phi = \nabla \cdot \left( \frac{3 \chi \kappa \varepsilon_0}{2} \nabla \phi \right) \tag{S14} \]

\[ \psi = -\nabla \cdot h^2 \nabla \phi + ((\phi^2 - 1)\phi \tag{S15} \]

\[ 2 \frac{\partial E_l}{\partial \phi} \right) \]

\[ F_e = \varepsilon_0 \varepsilon_r \left| E \right| \frac{\partial E}{\partial \phi} \tag{S19} \]

\[ F_e = -\frac{\partial \Delta E_l}{\partial \phi} \tag{S20} \]

where \( u \) denotes the velocity, \( \rho \) is the density, \( \mu \) is the dynamic viscosity, \( p \) is the pressure, \( I \) is the unit of the direction vector, \( g \) is gravity, \( F_i \) is the surface force, \( F_e \) is the divergence of the Maxwell stress tensor, \( \phi \) is the phase field variable, which is -1 in pollution and 1 in air, \( \kappa \) is a numerical parameter that determines the thickness of the fluid interface, and \( \chi \) is the mobility parameter of the interface.

**The stress analysis**

For pollution on superhydrophobic surfaces, there are three main forces in the electric field environment: surface force, Maxwell stress and interfacial external force (SI: N m\(^{-2}\)). The expression is shown as follows.

\[ F_s = \frac{3}{2\sqrt{2}} \kappa \gamma_n \frac{\partial \psi}{\partial \phi} \tag{S17} \]

\[ \psi_0 = -\nabla \cdot \kappa^2 \nabla \phi + ((\phi^2 - 1)\phi \tag{S18} \]

\[ F_e = \varepsilon_0 \varepsilon_r \left| E \right| \frac{\partial E}{\partial \phi} \tag{S19} \]

\[ F_e = -\frac{\partial \Delta E_l}{\partial \phi} \tag{S20} \]

where \( F_i \) is the surface force, \( \kappa \) is a numerical parameter that determines the thickness of the fluid interface, \( \gamma_n \) is the surface tension, \( F_e \) is the divergence of the Maxwell stress tensor, \( \varepsilon_0 \) is the dielectric constant of vacuum, \( \varepsilon_r \) is the dielectric constant of pollution, \( F_i \) is the interfacial external force, and \( \phi \) and \( \psi_0 \) are the phase field variables.

The basic variables of the phase field need to be involved in the further analysis of pollution dynamics. Here, we explain the meaning of related variables in detail.

Phase field variable \( \phi \)

The phase field variable \( \phi \) is the distinction pollution, external air and interface layer. The phase field variable \( \phi \) is -1 in pollution and 1 in air. There is an area of transition from the external air to the internal material, which is called the interface layer between the pollution and air. In this area, the phase variable is linearly distributed from 1 in air to -1 in pollution, as shown in Fig. S7. Furthermore, the interface layer thickness is assumed to be \( \kappa \) for the convenience of calculation.

*Figure S7* Schematic phase field variable \( \phi \). The value is -1 in pollution and 1 in air. \( \kappa \) is the interface layer thickness.

Phase field variable \( \psi_0 \)

The phase field variable \( \psi_0 \) is the parameter of the phase field change and the gradient function of the phase field variable \( \phi \) is shown in equation (S18). Due to the influence of the interface change and the elastic deformation energy, the influence of energy transformation on phase field variables is considered, as shown in Fig. S8. The improved equation is as follows:

\[ \psi = -\nabla \cdot \kappa^2 \nabla \phi + ((\phi^2 - 1)\phi + \frac{2\sqrt{2}\kappa}{3\gamma_n} \frac{\partial \Delta E_l}{\partial \phi} \tag{S21} \]

where \( \Delta E_l \) is the change in the system energy.
Figure S8 Schematic phase field variable $\psi_0$. (a) The initial value of the phase field variable $\psi_0$, which does not consider the change in system energy. (b) The initial value of the phase field variable $\psi_s$, which is considered the change in system energy. (c) At the jump moment, the value of the phase field variable $\psi_s$ which is not considered the change of system energy. (d) At the jump moment, the value of the phase field variable $\psi_{ij}$ does not consider the change in system energy. The change in system energy is different in the left and right regions.

In Fig. S8, the phase field variable $\psi_0$ is a key variable that significantly affects the aggregation and movement of pollution on the superhydrophobic surface. However, the change in system energy has a great influence on the imbalance in the process of defilement aggregation, which can lead to the force changing under the electric field. The jumping of this state is different from the equilibrium state. Therefore, in order to reveal the real motion characteristics, the non-equilibrium motion processes with the change in system energy are considered in the simulation analysis in this paper.

The relationship between the dynamics characteristics and electric field intensity

Combining equations (S10), (S17), (S19), (S20) and (S21), the relationship between the force and electric field intensity can be obtained as follows:

$$F_x = E^3 \frac{3\varepsilon_0 \varepsilon_r \kappa}{2\sqrt{2} \beta} \frac{\partial \psi_0}{\partial \phi} + \frac{3}{2\sqrt{2}} \kappa \gamma_0 \frac{\partial \psi_s}{\partial \phi} \quad (S22)$$

$$F_y = \epsilon_0 \varepsilon_r \vert F \vert \frac{\partial \bar{n}}{\partial \phi} \quad (S23)$$

$$\bar{n} = \frac{E}{|E|} \quad (S24)$$

$$F_x = E^2 \frac{\epsilon_0 \varepsilon_r \kappa}{2} \frac{\partial}{\partial \phi} (L + h - \pi R) + \gamma_0 \frac{\partial}{\partial \phi} (2L + 2h - 2\pi R) - k_D \frac{\partial}{\partial \phi} \frac{L}{R^2} \quad (S25)$$

where $\bar{n}$ is the unit vector in the direction of the electric field intensity.

From the above equations, it can be seen that the force on the pollution surface is proportional to the square of the electric field intensity. Therefore, the resultant force is simplified. The simplified equations are as follows:

$$F_r = \frac{\partial \bar{m}}{\partial \phi} (k_1 E^2 + k_2) \quad (S26)$$

$$\bar{m} = \frac{F_r}{|F_r|} \quad (S27)$$

where $F_r$ is the resultant force, $k_1$ is the quadratic term coefficient, $k_2$ is the constant coefficient, and $\bar{m}$ is the unit vector in the direction of the resultant force.

According to Newton's law, the relationship between the acceleration of the surface change and the surface resultant force is as follows:

$$a = \frac{F_r}{\rho} = \frac{\partial \bar{m}}{\partial \phi} \frac{(k_1 E^2 + k_2)}{\rho} \quad (S28)$$

where $a$ is the acceleration of the surface change, and $\rho$ is the pollution density.

The change states of pollution are similar during the self-aggregation process. It is assumed that the surface distance change in
pollution remains unchanged throughout the self-aggregation process. The relationship between the time and the total distance of the system is as follows:

$$\Delta S = \frac{1}{2} a t^2$$  \hspace{1cm} (S29)

where $\Delta S$ is the change in the surface distance, which is a constant, and $t$ is the self-aggregation process time.

The self-aggregation process time can be described by the resultant force as well as the electric field intensity. The equations are as follows:

$$t = \sqrt{\frac{2\Delta S}{a}} = \sqrt{\frac{2\Delta S \rho}{F_r}} = \sqrt{\frac{2\Delta S \rho}{(k_1 E^2 + k_2) \frac{\partial \phi}{\partial n}}}$$  \hspace{1cm} (S30)

Combining equations (S29) and (S30), the self-aggregation process time is inversely proportional to the electric field intensity.

**Note S3: Statistical analysis of the test results.**

In the experiment, the aggregation characteristics in different humidity environments were statistically analysed, as shown in Fig. S3. The humidity affects the aggregation time. With increasing humidity, the aggregation time is reduced in the same electric field. In the region with low humidity, the change in aggregation time is relatively slow, but in the region with high humidity, the change is obvious. A high humidity environment can effectively promote the self-aggregation of pollution as well as the self-cleaning. When the humidity is kept constant and the temperature is changed, the aggregation time changes slightly. This result shows that temperature has relatively little influence on the self-aggregation behaviour of pollution. To further research the relationship between the aggregation time and electric field intensity, the experimental data were fitted by statistical methods, as shown in Fig. S3. Considering the analysis results of Note S2, the experimental data should satisfy the following equation:

$$t(E) = n_1 E^{-1} + n_2$$  \hspace{1cm} (S31)

where $t(E)$ is a function of time and the electric field, $n_1$ is a quadratic fitting coefficient, and $n_2$ is a constant term fitting coefficient.

The relevant fitting parameters are shown in the Table S4.

**Table S4** The fitting parameters of experiment data

| $5.0 \text{ g kg}^{-1}$ | $5.4 \text{ g kg}^{-1}$ | $6.0 \text{ g kg}^{-1}$ | $7.2 \text{ g kg}^{-1}$ | $10 \text{ g kg}^{-1}$ | $18.8 \text{ g kg}^{-1}$ |
|-------------------------|-------------------------|-------------------------|-------------------------|-------------------------|-------------------------|
| $n_1$                   | 17.92                   | 15.95                   | 15.83                   | 8.785                   | 16.01                   | 15.93                   |
| $n_2$                   | 4.484                   | 3.925                   | 2.465                   | 0.525                   | 3.823                   | 3.777                   |
| RMSE                    | 0.6978                  | 0.5393                  | 1.1066                  | 0.8445                  | 0.4965                  | 0.4389                  |
| $R^2$                   | 0.9603                  | 0.9656                  | 0.9379                  | 0.8698                  | 0.9667                  | 0.9678                  |

where RMSE is the root-mean-square error.

Table S4 shows excellent fitting accuracy, and the experimental data show that the aggregation time is inversely proportional to the electric field intensity, which is similar to the theoretical analysis results. However, with increasing humidity, the fitting accuracy clearly decreases. We believe that the theory only considers the influence of the electric field, but does not consider the influence caused by the change in the water content in the pollution. In the actual experiment, it is found that the moisture absorption rate of the pollution is different in different environments, as shown in the Fig. S4. In a low humidity environment, the moisture absorption rate changes slightly. In a high humidity environment, the moisture absorption rate is significantly accelerated. The higher the humidity is, the faster the moisture absorption rate change will be and the more uneven the overall pollution will be. At this time, the uneven of pollution in the process of pollution aggregation cannot be ignored. In the original theory, the fixed parameter exhibits a great dynamic change, which leads to some errors in the calculation results.

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