Role of surface physicochemical properties of pipe materials on bio-clogging in leachate collection systems from a thermodynamic perspective

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Submitted to

Science of the Total Environment

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Abstract

Bio-clogging in pipes poses a significant threat to the operation of leachate collection systems. Bio-clogging formation is influenced by the pipe materials. However, the relationship between bio-clogging and the physicochemical properties of different pipe materials has not been clarified yet, especially from a thermodynamic aspect. In this study, the dynamic bio-clogging processes in pipes of different materials (high-density polyethylene (HDPE), polyvinyl chloride (PVC), polypropylene (PP), and polyethylene (PE)) were compared, and their correlation with the physicochemical properties was investigated. Results showed that the bio-clogging in HDPE and PVC pipes was more severe than that in PP and PE pipes. In bio-clogging development, the predominant factor changed from the surface roughness to the electron donator parameter (γ^-). In the initial phase, the most severe bio-clogging was observed in the HDPE pipe, which exhibited the highest roughness (432±76 nm). In the later phase, the highest γ^- (2.2 mJ/m^2) and protein content (2623.1±33.2 μg/cm^2) were observed in the PVC simultaneously. Moreover, the interaction energy indicated that the bacteria could irreversibly and reversibly adhere to the HDPE, whereas irreversible adhesion was observed in the PVC, PP, and PE cases. The findings clarify the thermodynamic mechanism underlying bio-clogging behaviors and provide novel insights into the bio-clogging behaviors in pipes of different materials, which can facilitate the development of effective bio-clogging control strategies.

Keywords: Bio-clogging, Pipe materials, Leachate, Thermodynamic analysis, Interaction energy

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1. Introduction

The clogging of leachate collection systems (LCSs) represents a collective environmental challenge in landfills. As a key component of LCSs, pipes are commonly used to collect and transport leachate to treatment facilities (Shaha et al., 2019). Different materials, such as high-density polyethylene (HDPE), polyvinyl chloride (PVC), polypropylene (PP), and polyethylene (PE), have been used to prepare pipes (Tarek and Daria, 2015). Many fields and laboratory studies have indicated varying degrees of bio-clogging in pipes made of different materials (Liu and Liu, 2021; Rowe and Yu, 2012; Tang et al., 2018). For instance, Li et al. (2021) reported that the bacterial amount related to bio-clogging in PVC was higher than that in HDPE. In general, bio-clogging decreases the hydraulic conductivity of pipes and leachate flow rate, which increases the possibility of LCSs clogging. LCSs clogging poses a threat to landfill stability and may lead to the pollution of the surrounding environment (Feng et al., 2020; Wu et al., 2018). Therefore, exploring the bio-clogging mechanisms related to different pipe materials in LCSs is critically important to controlling clogging in landfills.

Although many researchers have focused on the relationship between bio-clogging and physicochemical properties (e.g. surface free energy, Zeta potential, interaction energy) of different pipe materials in domestic wastewater treatment plants, the bio-clogging mechanisms in landfills have not been extensively investigated. Zhang et al. (2019) noted that in wastewater treatment plants, the surface free energy and Zeta potential could influence bacteria adhesion behaviors in different pipe materials. Additionally, the interaction energy of pipe materials could influence the secretion of

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protein and polysaccharides, which represent the main components of extracellular polymeric substances (EPSs) (Teng et al., 2022). Both bacteria adhesion and EPSs secretion are key processes in bio-clogging development (Carrel et al., 2018; Wang et al., 2021b). Notably, the bio-clogging characteristics are influenced by not only the pipe materials but also the type of wastewaters (e.g. sewage, leachate). Leachate is a type of heterogeneous wastewater with various compositions and contains high-level pollutants, i.e., bacteria, organic matter, and inorganic ions, which render the bio-clogging phenomena complex (Luo et al., 2020; Anand et al., 2022). Our previous study illustrated that the leachate characteristics could change the bio-clogging components and microbial communities (Wang et al., 2021b). Hence, the relationship between bio-clogging and the physicochemical properties of pipe materials exposed to leachate must be explored to facilitate the control of bio-clogging behaviors pertaining to leachate.

Recently, different thermodynamic theories have been used to describe and predict the adhesion behaviors between bio-clogging constituents and material surfaces (Zeng et al., 2022; Wu et al., 2020). The extended Derjaguin–Landau–Verwey–Overbeek (XDLVO) theory has been regarded as the ideal theory for describing the bio-clogging-related adhesion behaviors related to materials encountered in domestic wastewater treatment (Habimana et al., 2014; Yin et al., 2020). The XDLVO theory is based on acid-based, van der Waals, and electrostatic double-layer interaction energies (Somathilake and Hettiaratchi, 2012). Based on the XDLVO theory, Yongabi et al. (2020) reported that the adhesive behavior between bacteria and the material surface is controlled by the electrostatic double-layer interaction energy at the initial contact,

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whereas stable adhesion is driven by the van der Waals energy. In addition, the XDLVO theory has been to interpret the dynamic adhesion behavior of protein and polysaccharides on the membrane surface in domestic wastewater treatment (Teng et al., 2022). However, to the best of the authors' knowledge, little research has been conducted to identify the relationship between the bio-clogging and physicochemical properties of pipe materials subjected to leachate, especially considering the influence of the leachate dynamic characteristics on the dynamic evolution of bio-clogging.

Therefore, this study was aimed at clarifying the influence of the physicochemical properties of different pipe materials subjected to leachate on the dynamic evolution of bio-clogging and the related thermodynamic mechanisms. Experimental characterizations based on confocal laser scanning microscopy (CLSM), colony-forming unit (CFU) measurements, and 16S rRNA gene sequencing were performed to characterize the dynamic changes in bio-clogging. The physicochemical properties (roughness, hardness, Zeta potential, and surface energy) of four common kinds of pipe materials were compared. Moreover, the XDLVO theory and redundancy analysis (RDA) was used to describe the effects of physicochemical properties on bio-clogging. The findings can provide novel thermodynamic insights into leachate-related bio-clogging, thereby promoting the development of bio-clogging control strategies.

2. Material and methods

2.1 Pipe materials and chemicals

Four most commonly used pipe materials, i.e., HDPE, PVC, PP, and PE, were considered in this study (Alimi et al., 2018). HDPE, PVC, PP, and PE were purchased

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from markets in Shenzhen, China. With reference to a previous study (Cai et al., 2019), the pipe materials were cut into 1.0×1.0 cm squares using sterile scissors, washed with ethanol and ultrapure water two times, and air-dried for further analyses. As illustrated in Fig. S1, the FTIR spectra of these four pipe materials are compared. All the chemicals were purchased from Xilong Scientific (Shantou, China). Unless specified, all chemicals were dissolved in ultrapure water obtained from a Milli-Q ultrapure water purification system (Millipore, USA).

2.2 Characterizations of pipe materials

The physicochemical surface properties of the pipes, including the roughness, hardness, Zeta potential, and contact angle were analyzed. The pipe roughness was determined through atomic force microscopy (AFM) (Dimension Icon, Bruker, USA) with a scan range of 50.0×50.0 μm, and a scanning speed of 0.4-0.6 Hz. Specifically, AFM images were analyzed using the NanoScope Analysis software to calculate the roughness. Measurements for each pipe material were conducted in triplicate. The pipe hardness was determined using a Shore durometer (LXD-D, Shanghai Siwei Instrument Manufacturing Co., China) according to the ASTM D2240D standard (Cai et al., 2019). For each type of pipe material, measurements were obtained for at least six sites. The Zeta potentials of the pipe materials and leachate were measured using a Zetasizer (Nano ZS, Malvern, UK), based on the electrophoretic light scattering spectrophotometer method. The pipe materials were crushed and grinded as pretreatment for the Zeta potential analysis. Each measurement was performed at least six times at ambient temperature (25±2℃). The major functional groups of pipe

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materials were determined through Fourier transform infrared spectroscopy (FTIR) (IRTracer-100, Shimadzu, Japan) between 400 cm^{-1} and 3600 cm^{-1} (Fig. S1).

The contact angle of the probe liquid was measured using a contact angle meter, based on the drip-stop method (ThetaLite, Biolin Attention, Sweden). Ultrapure water, diiodomethane, and ethylene glycol were selected as the probe liquids to derive the surface tension components of the solids. The contact angle for one pipe material was measured based on five samples. The contact angle for the bacteria was measured through bacterial lawns, obtained by filtering the leachate through a Millipore filter (0.22 μm) (Zhang et al., 2019). The total surface tension γ contained two components: an apolar Lifshitz-van der Waals component, γ^{LW}; and a polar acid-based component, γ^{AB}. The acid-based component consisted of an electron acceptor (γ^{+}) and electron donator (γ^{-}). Therefore, the total surface tension of the pipe material and bacteria was calculated as follows (Liu et al., 2007; Van Oss, 1994):

$$\gamma = \gamma^{LW} + \gamma^{AB} = \gamma^{LW} + 2\sqrt{\gamma^{+}\gamma^{-}}$$

(1)

$$(1 + \cos\theta)\gamma_L = 2\left(\sqrt{\gamma_S^{LW}\gamma_L^{LW}} + 2\sqrt{\gamma_S^{+}\gamma_L^{-}} + \sqrt{\gamma_L^{+}\gamma_S^{-}}\right)$$

(2)

Where θ is the contact angle of three polar liquids (ultrapure water, diiodomethane, and ethylene glycol). γ_L is the Lifshitz-van der Waals, electron acceptor, and electron donator parameters of different probe liquids (Table S1). γ_S^{LW} and γ_L^{LW} represent the Lifshitz-van der Waals parameter of the pipe material surface and probe liquid, respectively. γ_S^{+} and γ_L^{+} represent the electron acceptor parameter

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of the pipe surface and probe liquid, respectively. γ_S^- and γ_L^- represent the electron donator parameter of the pipe surface and probe liquid, respectively.

2.3 Preparation of bacterial strains

The bacterial strains and leachate were collected from the simulated municipal solid waste (MSW) landfill reactor. The synthetic MSW was composed of 65% food waste, 10% paper, 10% sand, 10% pipe material, and 5% others (glass and metals). The pH, conductivity, and ammonia concentration of leachate were 5.28, 6.65 mS/cm, and 460 mg/L, respectively (Table S2). The community structures of leachate were detected by the 16 S rRNA with the primer sets of 338F (5'-ACTCCTACGGGAGGCAGCAG-3') and 806R (5'-GGACTACHVGGGTWTCT-3'). Polymerase chain reactions (PCR) reactions were performed in a mixture containing a buffer, primer, and template DNA. The PCR products were extracted from a 2% agarose gel and further purified and quantified. The purified amplicons were paired-end sequenced on an Illumina MiSeq platform according to the standard protocols specified by Majorbio Bio-Pharm Technology Co. Ltd.

2.4 Bio-clogging formation experiment

A bio-clogging formation experiment was performed in 200 mL beakers. The experimental groups contained 50 mL leachate and 6 pieces of sterilized pipe materials, whereas the control groups contained 50 mL sterilized ultrapure water and 6 pieces of sterilized pipe materials. Each beaker was flushed with nitrogen for 10 min and rapidly sealed with a frosted cover to maintain anaerobic conditions. Finally, all the beakers were incubated in a thermostat at 35±2℃. According to previous studies (Wang et al.,

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2021a; Wang et al., 2022), the pipe materials in the four beakers were extracted on days 1 and 14, representing phases I and II, respectively, for further analyses. Phase I corresponded to the initial phase of bio-clogging formation, mainly caused by bacteria adsorption (Tang et al., 2018). Phase II represented the later phase of bio-clogging formation, with increased EPS contents (Wang et al., 2022).

2.5 Analysis of bio-clogging characteristics

The bio-clogging materials were characterized in terms of morphology, component distribution, EPS contents, and bacterial contents. The morphology was analyzed by scanning electron microscopy (SEM) (Supra®55, Lecia, Germany) at 15 kV. The SEM samples were prepared according to a previous study (Wang et al., 2021b). Firstly, the pipe materials were fixed using a primary fixative (2.5% glutaraldehyde in 0.1 M phosphate buffer). Subsequently, the pipe materials were dehydrated with increasing concentrations of ethanol solutions (30%, 50%, 70%, 85%, 90%, and 100%) and substituted with isoamyl acetate. The samples were frozen at -20℃ and -80℃ for 4 h, respectively. Finally, the freeze-dried samples were sputter-coated with Au and imaged using SEM.

The distribution of the bio-clogging components was detected by CLSM (AIR, Nikon, Japan). Four pipe coupons were obtained from the clogged pipe sample by cutting with sterile scissors. The coupons were stained with SYTO 9 and propidium iodide (PI) for the detection of live cells and dead cells, respectively. The concentrations of SYTO 9 and PI were 3.34 and 20 μM, respectively. The stained coupons were incubated in the dark for 30 min and then rinsed with 0.1 M phosphate buffer solution

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(PBS) three times before imaging. The images were further analyzed using NIS-Element and Image J (National Institute of Health, USA).

The EPS was extracted by a heating method as previously described (Zhang et al., 2009). The pipe materials were put into 2 mL PBS and ultrasonic for 10 min, followed by heat treatment at 80°C for 30 min. The heat-treated samples were subjected to 10 min of ultrasonication and then centrifuged at 10000 rpm for 10 min (20°C). The polysaccharide and protein contents were detected using phenol-sulfuric acid and diquinoline formic acid methods, respectively (Dubois et al., 1956; Osnes et al., 1993).

The bacterial contents were measured using the CFU method. After the bio-clogging experiments, the pipe materials with attached bacteria were suspended and gently rinsed with PBS solution three times to remove the culture medium. For the remaining attached bacteria, the pipe materials were placed in 5 mL PBS, and the mixture was vigorously shaken manually and by using a vortex for 30 s. Finally, a 50 μL sample was dropped onto beef-extract peptone with a sterile pipette and incubated at 35±2°C for 24 h. The colonies on the beef-extract peptone plates were counted to determine the number of viable bacteria in each sample. Replicated experiments were performed for the colonies on the plates for each sample (n=4). Quantitative PCR (Q-PCR) was conducted to analyze the bacterial density using primer sets of 338F (5'-ACTCCTACGGGAGGCAGCAG-3')/806R (5'-GGACTACHVGGGTWTCTAAT-3'). The reactions were implemented using an ABI7300 real-time PCR system (Applied Biosystems, USA). The community structures of bio-clogging were detected as described in Section 2.3.

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2.6 Thermodynamic evaluation of the interaction energy

The Gibbs adhesion energy between the pipe materials and bacteria in leachate can be calculated using the XDLVO approach (Teng et al., 2019). The total adhesive energy primarily consists of Lifshitz-van der Waals energy, G_{adh}^{LW}, acid-based energy, G_{adh}^{AB}, and electrostatic double-layer interaction energy, $\Delta G_{\mathrm{adh}}^{EL}$, which can be calculated by (Wu et al., 2020; Xu et al., 2016):

$$\Delta G_{adh} = \Delta G_{adh}^{LW} + \Delta G_{adh}^{AB} + \Delta G_{adh}^{EL} \quad (3)$$

$$\Delta G_{adh}^{LW} = -2\left(\sqrt{\gamma_b^{LW}} - \sqrt{\gamma_l^{LW}}\right)\left(\sqrt{\gamma_S^{LW}} - \sqrt{\gamma_l^{LW}}\right) \quad (4)$$

$$\Delta G_{adh}^{AB} = 2\left(\sqrt{\gamma_b^{+}} - \sqrt{\gamma_S^{+}}\right)\left(\sqrt{\gamma_b^{-}} - \sqrt{\gamma_S^{-}}\right) - 2\left(\sqrt{\gamma_b^{+}} - \sqrt{\gamma_l^{+}}\right)\left(\sqrt{\gamma_b^{-}} - \sqrt{\gamma_l^{-}}\right) - 2\left(\sqrt{\gamma_S^{+}} - \sqrt{\gamma_l^{+}}\right)\left(\sqrt{\gamma_S^{-}} - \sqrt{\gamma_l^{-}}\right) \quad (5)$$

$$\Delta G_{adh}^{EL} = \frac{\varepsilon\kappa(\zeta_b^2 + \zeta_s^2)}{2}\left[1 - \coth \kappa d_0 + \frac{2\zeta_b\zeta_s}{\zeta_b^2 + \zeta_s^2}\operatorname{csch} \kappa d_0\right] \quad (6)$$

$$\kappa = \sqrt{\frac{4N_A e^2 I}{\varepsilon KT}} \quad (7)$$

Where subscript b, s, and l mean bacteria, pipe material, and leachate, respectively. ζ and κ mean Zeta potential (mV) and reciprocal Debye length (nm^{-1}), respectively. d_0 represents the minimum separation distance (nm) within two flat surfaces. N_A is the Avogadro constant, e is the elementary charge of an electron, I is ionic strength, K is the Boltzmann constant, ε is the dielectric constant, and T is temperature. The value of different parameters used in this study was presented in Table S3. In this study, 0.2 M and 0.158 nm were set as the ionic strength and minimum

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separation distance (Chen et al., 2012; Townsend, 2015). The Gibbs adhesion energy was calculated by the average values of different parameters.

The interaction energy between the bacteria and pipe materials was calculated assuming the entities as two surfaces. Similar assumptions have been used in the previous study (Teng et al., 2019). The XDLVO theory represents the primary method to calculate the interaction energy for the two surfaces (Van Oss, 1994). Accordingly, the total interaction energy (ΔG^{TOT}) can be calculated as follows:

$$\Delta G^{TOT}(\mathrm{d}) = \Delta G^{LW}(\mathrm{d}) + \Delta G^{EL}(\mathrm{d}) + \Delta G^{AB}(\mathrm{d}) \quad (8)$$

$$\Delta G^{LW}(\mathrm{d}) = \mathrm{S}\Delta G_{adh}^{LW}\frac{{d_0}^2}{d^2} \quad (9)$$

$$\Delta G^{AB}(\mathrm{d}) = \mathrm{S}\Delta G_{adh}^{AB}\exp\left(\frac{d_0 - d}{\lambda}\right) \quad (10)$$

$$\Delta G^{EL}(\mathrm{d}) = \mathrm{S}\varepsilon\kappa\zeta_b\zeta_s\left[\frac{(\zeta_b^2 + \zeta_s^2)}{2\zeta_b\zeta_s}(1 - \coth \kappa d) + \frac{1}{\sinh \kappa d}\right] \quad (11)$$

Where S is the interaction surface area between bacteria and pipe materials (m^2).

2.7 Statistical analysis

The results were expressed as mean ± standard deviation values. One-way analysis of variance was performed to evaluate the statistical differences. All the significance analyses were performed using the PASW 18.0 software (SPSS Inc., USA), and a p-value of <0.05 was considered to indicate statistical significance (Table S4-5). Redundancy analysis (RDA) was performed to clarify the relationship between the bio-clogging characteristics and pipe surface physicochemical properties using Canoco (Version 5.02, ScientiaPro, Hungary). A person's correlation between two parameters among different surface physicochemical properties and bio-clogging characteristics

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was calculated and compared.

3. Results and discussion

3.1 Characteristics and composition of bio-clogging

Fig. 1 shows the morphological characterization of different pipe materials. In terms of the bare pipe materials, the surfaces of HDPE and PVC were rough and coarse, whereas those of PP and PE were smooth. In phase I, the bacteria adsorbed on the surface of HDPE, PVC, and PP exhibited a rod-like and spherical structure with several attached bacteria and broken cells. Significant bacterial adhesion was not observed on the PE surface. The number of clogged materials in the four pipe materials considerably increased from phase I to phase II. The maximum amount of bacteria was observed on the HDPE surface, followed by the PVC and PP, and the PE exhibited the least amount of bacteria. These results indicated the clogging extent was associated with the pipe materials and phases, consistent with a previous study that highlighted that the clogging extent was positively correlated with the phase development (Ko et al., 2019). The bacteria could adhere to both rough and smooth pipe material surfaces by secreting EPSs to attach with the surface (Ganesan et al., 2022). Overall, the bacteria could develop bio-clogging on different pipe material surfaces with colonization ability, such as bacterial growth and EPSs secretion.

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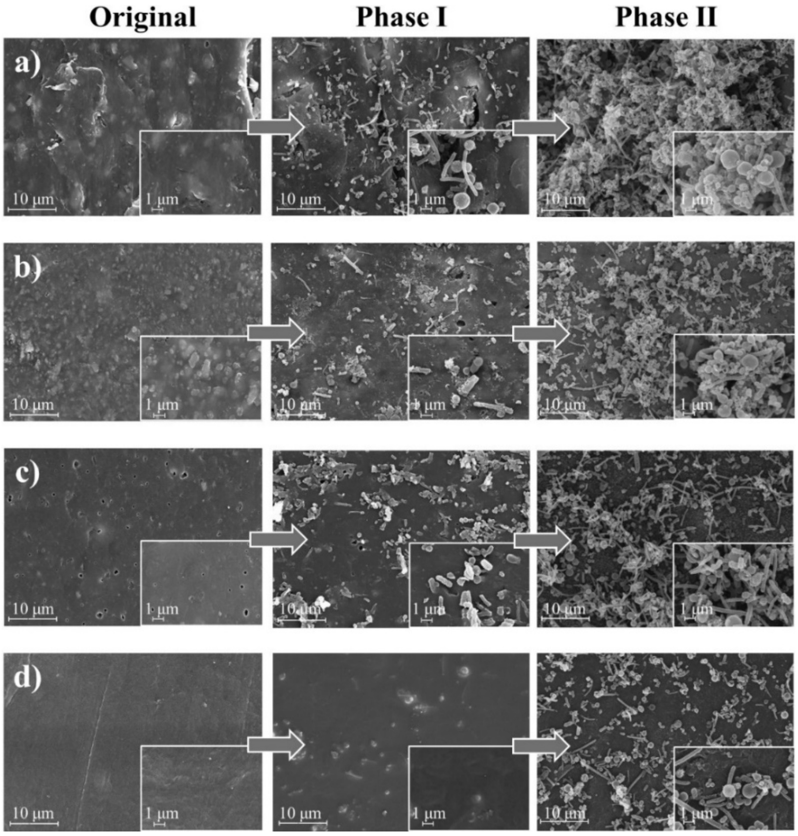

Fig. 1 Surface morphology of different pipe materials in different phases. a) HDPE, b) PVC, c) PP, d) PE.

As illustrated in Fig. 2, the bacteria density and EPS contents differed across the pipe materials and phases. In phase I, the maximum cell density corresponded to HDPE ($3.1\times10^5\pm0.6\times10^5$ CFU/cm^2), followed by PVC ($2.1\times10^4\pm0.7\times10^4$ CFU/cm^2), PP ($2.5\times10^3\pm0.3\times10^3$ CFU/cm^2), and PE ($3.3\times10^2\pm1.2\times10^2$ CFU/cm^2). Similar trends were observed in phase II, although the cell density for the different pipe materials in phase II was 1–2 orders of magnitude lower than those in phase I. In contrast, the total EPSs content increased from phase I to phase II. In phase I, the EPS content ranged from 199.7±9.1 to 583.4±11.1 μg/cm^2. Notably, EPSs were the main component of bio-clogging in phase II, with contents ranging from 973.8±51.6 to 2642.6±36.2 μg/cm^2. These results suggested that the protein and polysaccharides contents increased with

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the increasing phase, and the protein content was higher than that of the polysaccharides. The proteins, secreted by bacteria, aggregated in response to the bacterial adhesion to form EPSs and mediate the bacterial contacts, leading to an increase in the thickness of bio-clogging (Flemming and Wingender, 2010). In addition, the EPSs secretion and bacterial adhesion were associated with the physicochemical properties of the pipe surface, such as the roughness, charge, and interaction energy (Teng et al., 2019). For example, the highest EPSs contents were observed in PVC, whereas the HDPE corresponded to the highest cell density. The cell density of PE did not change considerably, although the cell densities of HDPE, PVC, and PP decreased from phase I to phase II. These results also indicated that the physicochemical properties of pipe materials could change microbial activities.

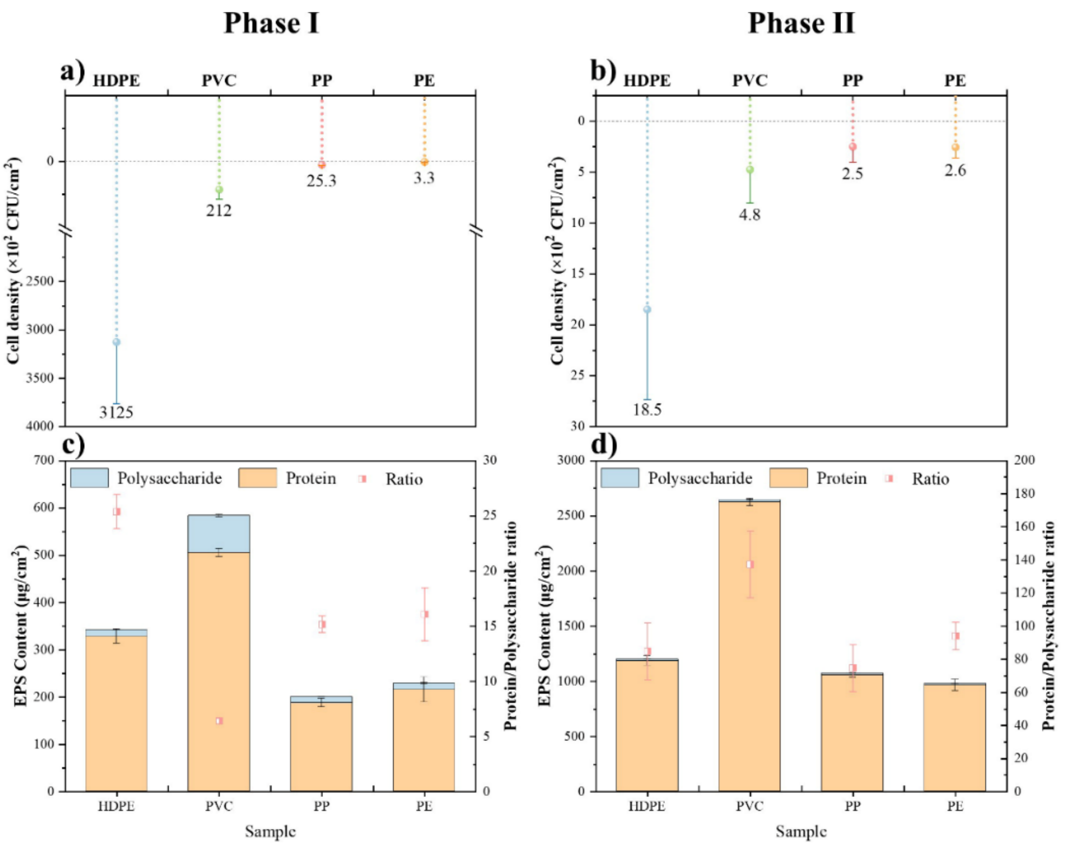

Fig. 2 Attached cell density for different pipe materials. a) phase I, b) phase II. EPS contents and

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protein/ polysaccharide ratios of different pipe materials. c) phase I, d) phase II.

Fig. 3 shows the fluorescent staining results of live and dead bacteria identified from different pipe materials. The live and dead bacteria were unevenly distributed on the surface of pipe materials (Fig. 3a, b). In phase I, the live bacteria (green) were the main contributors to bio-clogging, accounting for 54.0±3.8 to 77.8±0.6% of the total content (Fig. 3c). The percentage of live cells in the HDPE (77.8±0.6%) was the highest, consistent with the highest cell density observed for the HDPE (Fig. 2a). In contrast, a highly dense red assembly was observed in phase II, suggesting the dominant presence of dead bacteria (Fig. 3d). The main component of bio-clogging changed from live bacteria to dead bacteria and EPSs in the bio-clogging development process (Meng et al., 2017). The percentage of live bacteria (Phase I) exhibited the following decreasing order: HDPE>PVC>PE>PP. The percentage of dead bacteria the phase I exhibited the following decreasing order: PP>HDPE>PVC>PE. These results highlighted that the physicochemical properties of pipe materials changed the distribution of bio-clogging under the same phase. Fig. S2 shows the bacterial communities in different pipe materials. In phase I, the main bacteria in the HDPE, PVC, and PP cases were *Proteobacteria* (57.1%), *Firmicutes* (46.4%), and *Actinobacteria* (40.4%), respectively. The bacteria in the case of PE mainly consisted of *Firmicutes* (34.2%) and *Actinobacteria* (32.9%). In other words, *Proteobacteria*, *Firmicutes*, and *Actinobacteria* exhibited strong adaptability and broad ubiquity in the bio-clogging and were abundantly present in the leachate (Xu et al., 2017). The physicochemical properties of the different pipe materials changed the bacterial communities in the bio-

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clogging (Yan et al., 2022). The predominant bacteria for different pipe materials changed from phase I to phase II. For example, the main bacteria for HDPE in phase II were *Actinobacteria* (35.0%). In addition, a significant change was not observed in the leachate characteristics among different pipe materials (Table S6), suggesting the pipe material could not change the leachate characteristics and bio-clogging phases. Overall, these results indicated that the introduction of differences in the species across bio-clogging was a dynamic process related to the pipe materials and bio-clogging phases.

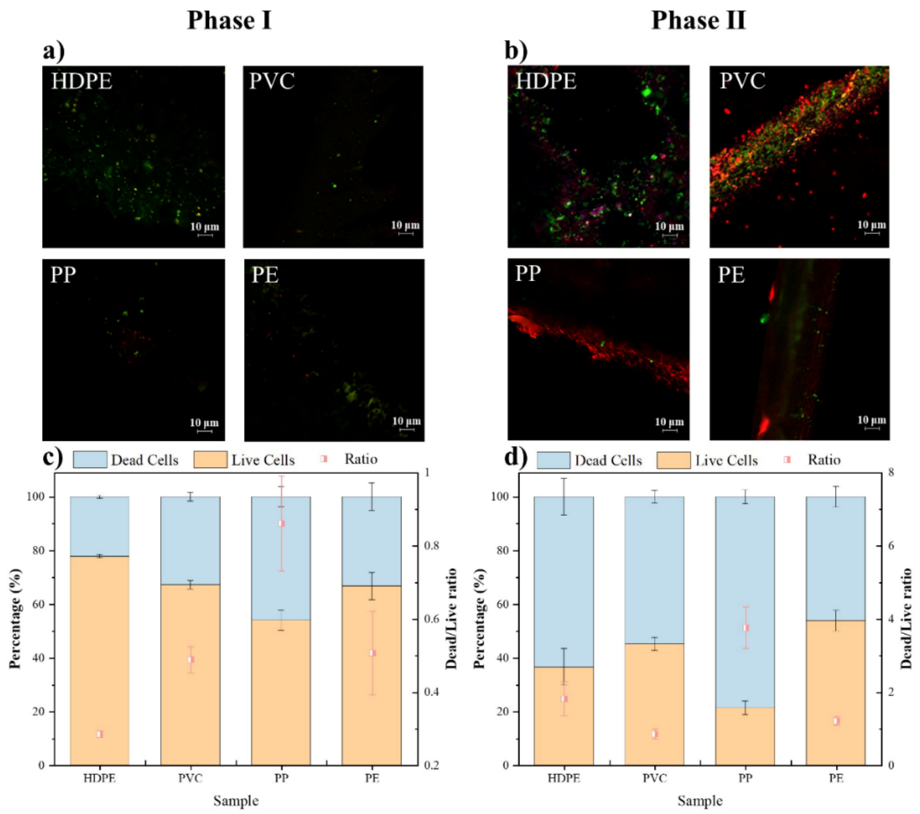

Fig. 3 Representative CLSM images of bio-clogging in different pipe materials. a) phase I, b) phase II. Live cells (green), dead cells (red). Distribution of live cells and dead cells in different pipe materials. c) phase I, d) phase II.

3.2 Effects of physicochemical properties on the bio-clogging

The morphology and roughness of the pipe materials were visualized through

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AFM (Fig. 4). As illustrated in Fig. 4, HDPE exhibited the highest average roughness (432±76 nm), followed by the PVC (190±15 nm), PE (149±19 nm), and PP (69±5 nm). The roughness influenced the contact area between the bacteria and the surface, thereby affecting the bacterial adhesion (Yang et al., 2022). Notably, although the roughness of PE was higher than that of PP, the less bio-clogging formation was observed in PE compared to that in PP (Fig. 1 and 2). Thus, the surface roughness likely did not represent the main factor for the bio-clogging formation in different pipe materials. This observation is also supported by a previous study that highlighted that nanoscale surface roughness could not provide micro crevices that could function as niches for bacteria (Sousa et al., 2009). However, several recent studies have indicated that a high roughness can influence the interaction force between bio-clogging (bacteria) and the interface surface and decrease the interaction energy barrier between the two interacting bodies (Bradford et al., 2018), thereby generating more favorable conditions for the interaction. Thus, in this study, the effect of roughness on bio-clogging formation was further evaluated, as described in Section 3.3.

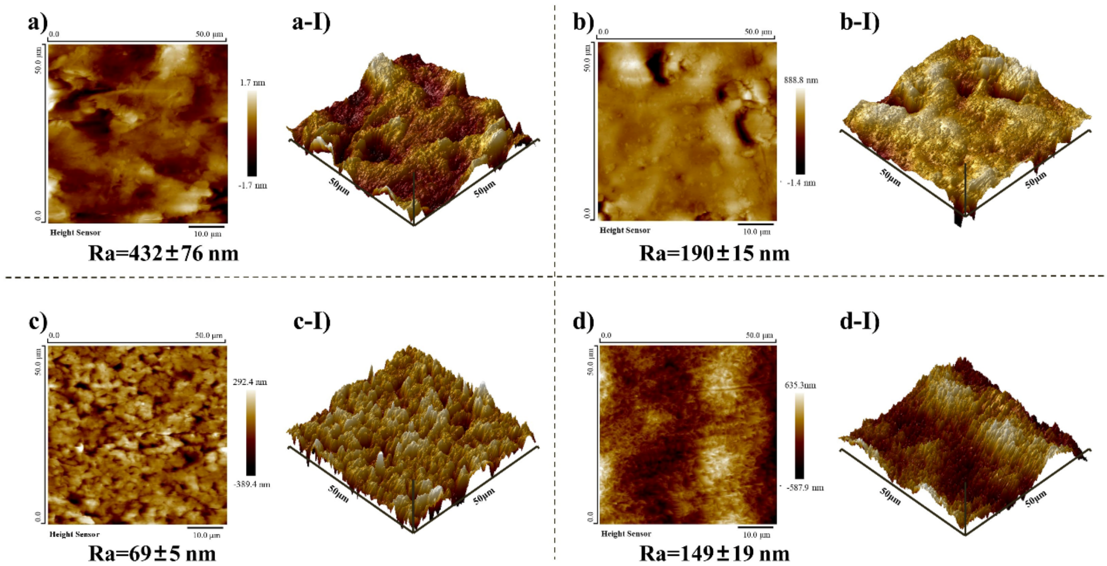

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Fig. 4 AFM deflection images and adhesion force measurements for different pipe materials. a) HDPE, b) PVC, c) PP, d) PE.

The surface hardness of the pipe materials could influence bacterial adhesion because hard semi-solid surfaces are expected to limit bacterial swarming (Kamatkar and Shrout, 2011; Cai et al., 2019). As illustrated in Fig. S3a), the surface hardness of HDPE (64.0±0.5 HD) and PVC (82.8±0.5 HD) was lower than those of PE (96.2±0.4 HD) and PP (98.4±0.2 HD). PE and PP exhibited lower bacterial adhesion than HDPE and PVC. This finding indicated that the amount of bio-clogging formation was positively correlated with the surface hardness, consistent with previously reported findings (Cai et al., 2019). Moreover, the surface charge could affect bio-clogging formation through electrostatic interactions (Yang et al., 2022). The HDPE exhibited a significant negative charge, with a Zeta potential of -21.2±0.3 mV (Fig. S3b). The differences in the Zeta potential of PP and PE were not statistically significant ($P>0.05$). The bacteria used in this study had a Zeta potential of -7.8±0.8 mV. These results indicated that the electrostatic interaction does not considerably influence the bio-clogging formation. To clarify the bio-clogging formation behavior between the bacteria and pipe materials, the thermodynamic approach and XDLVO theory were used to evaluate the contact angle, surface free energy, and Gibbs adhesion energy.

The contact angles were measured to investigate the hydrophilicity/ hydrophobicity of the pipe materials (Fig. 5). As illustrated in Fig. 5a, the water contact angle of all pipe materials was more than 90°, suggesting that all pipe materials were hydrophobic. Notably, the maximum water contact angle corresponded to HDPE

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(111.2±2.7° , P<0.01). Similar trends were also observed in other probe liquids, including diiodomethane and ethylene glycol. These contact angles were used to calculate the surface free energy of the pipe materials. As illustrated in Figure 5b, the minimum total surface energy (γ) corresponded to HDPE (19.66 mJ/m^2), followed by PP (27.23 mJ/m^2), PE (31.95 mJ/m^2), and PVC (35.49 mJ/m^2). The Lifshitz–van der Waals parameter was noted to be the main contributor to the total surface energy for different pipe materials. These results indicated that HDPE corresponded to the lowest surface energy and most severe bio-clogging formation. Notably, our results were in contrast with those of previous studies that indicated that the bacteria adhesion was positively correlated with the surface energy (Merghni et al., 2016; Mohiuddin et al., 2011). As mentioned previously, HDPE corresponded to the highest roughness (Fig. 4). In general, for hydrophobic pipe materials, a larger contact angle corresponds to a rougher surface and more functional groups (Niu et al., 2021). In this context, the HDPE contained more carbonyl groups, as confirmed by the result of FTIR at wavelengths of 1600–1900 cm^{-1} (Fig. S1) (Syranidou et al., 2017). Overall, bio-clogging formation is a complex process influenced by different physicochemical properties, such as roughness, hardness, surface charge, and hydrophilicity. However, the effect and mechanisms of hydrophobicity and surface free energy of pipe materials on the bio-clogging need to be extensively investigated.

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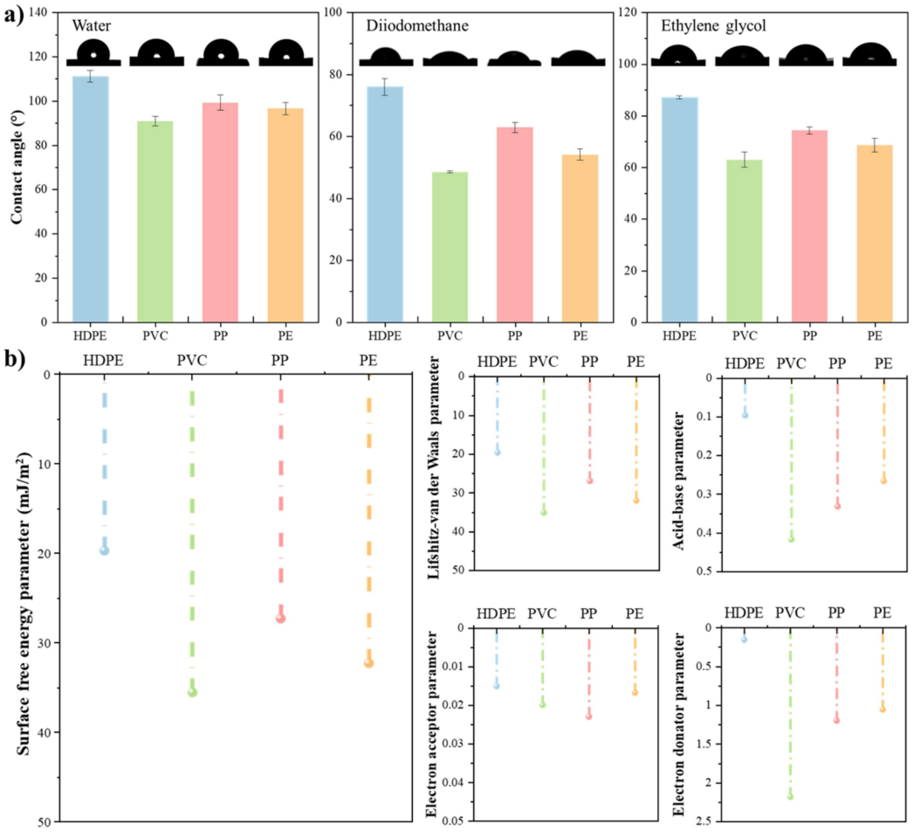

Fig. 5 a) Contact angles of different pipe materials in three probe liquids (water, diiodomethane, ethylene glycol). b) Surface free energy of different pipe materials, in terms of the Lifshitz-van der Waals, acid-base, electron acceptor, and electron donator parameters. The unit for all parameters is mJ/m^2.

3.3 The interaction energy between the pipe materials and bio-clogging

Fig. 6 illustrates the dependence of the interaction energy between bacteria and different pipe surfaces on the separation distance. The total interaction energy for the four pipe materials decreased with the increase in the separation distance. In general, the total interaction energy at minimum separation distance (d_0=0.158 nm) ($G_{d_0}^{TOT}$) is a critical index of the adhesive ability of bacteria (Chen et al., 2012; Wu et al., 2020). The $G_{d_0}^{TOT}$ of HDPE, PVC, PP, and PE were 2.5×10^{14}, 5.2×10^{14}, 4.0×10^{14}, and 4.5×10^{14} KT, respectively. These results suggested that the bacteria preferentially adhered

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to the HDPE owing to the lower energy barrier, consistent with the experimental observation (Fig. 2). The high roughness decreased the interfacial energy and promoted bio-clogging attachment (Zhao et al., 2015). Therefore, the bacteria could overcome the lowest energy barrier (2.5×10^{14} KT) in the HDPE to exhibit irreversible adhesion, although a larger amount of bacteria adhered reversibly as they preferred to overcome the lower energy barrier with the minimum expended energy (about 62.0 KT). For the PVC, PP, and PE, a primary minimum expended energy was observed, suggesting that more bacteria could adhere irreversibly.

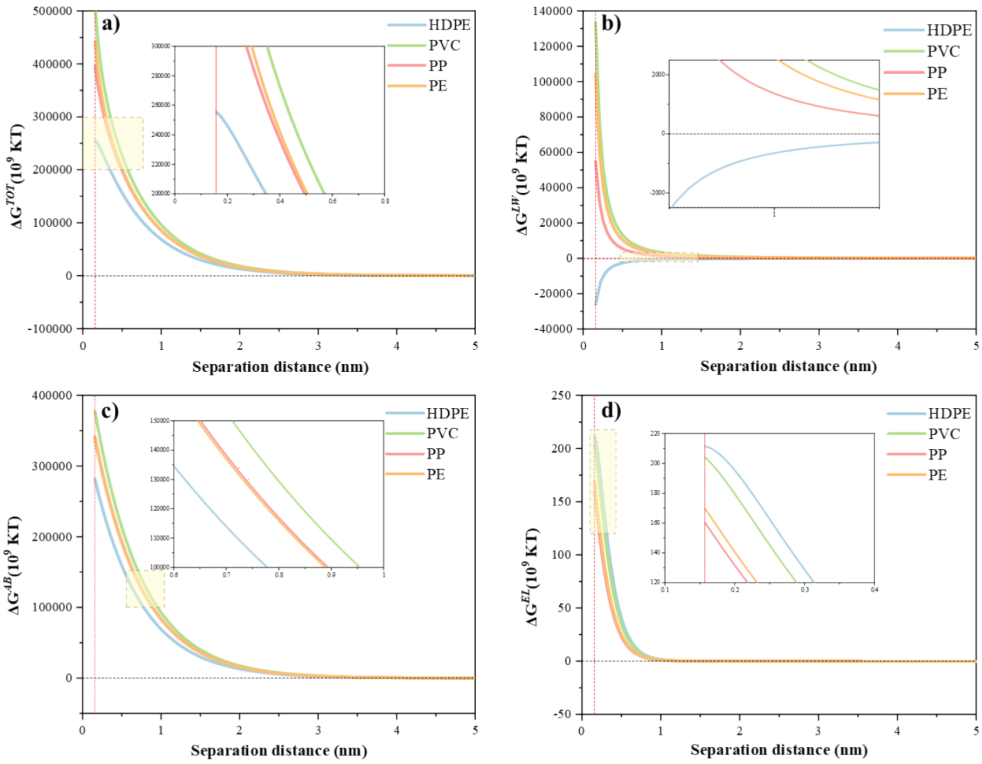

Fig. 6 Interaction energies between bacteria and pipe materials, obtained from the XDLVO analysis of a) total interaction energy, b) van der Waals energy, c) acid-base interaction energy, and d) electrical double-layer energy.

To clarify the influence of different forces on the total interaction energy, the Lifshitz–van der Waals (ΔG^{LW}), acid-base (ΔG^{AB}), and electrostatic (ΔG^{EL}) interactions

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for the different materials were evaluated, as shown in Figs. 6b–d, respectively. ΔG^{LW} was determined by the Hamaker constant. The Hamaker constants of HDPE, PVC, PP, and PE were -1.08×10^{-7}, 5.52×10^{-7}, 2.28×10^{-7}, and 4.33×10^{-7} J, respectively (Table S7). According to Eq. (9), the ΔG^{LW} of the HDPE was negative, whereas the ΔG^{LW} values of PVC, PP, and PE were positive. These results illustrated that strong Lifshitz–van der Waals attractive forces existed between the bacteria and HDPE, whereas repulsive effects were observed between the bacteria and other pipe materials (PVC, PP, and PE). Moreover, the ΔG^{AB} of the different pipe materials were similar. ΔG^{AB} exhibited the maximum energy barriers in different pipe materials, compared to ΔG^{LW} and ΔG^{EL}. These results suggested that repulsive Lewis acid-base interactions occurred between the bacteria and different pipe materials. In addition, the ΔG^{EL} of different pipe materials exhibited similar trends, owing to the comparable Zeta potentials of different pipe materials (Fig. S3b). Thus, the electrostatic interaction did not considerably influence the bacterial adhesion. These results were consistent with a previous study that highlighted the absence of any significant differences in ΔG^{EL} for different pipe materials (Cai et al., 2019). Overall, the consistency of the experimental observations with the thermodynamic prediction based on the XDLVO method demonstrated the feasibility of this approach.

3.4 Comprehensive analysis of the relation between bio-clogging and physicochemical properties

The relationships between the bio-clogging factors and pipe material physicochemical properties in phase I and phase II were revealed by RDA (Fig. 7). In

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phase I, RDA1 and RDA2 accounted for 65.0% and 28.3% of the bio-clogging differences in the groups, respectively. The first two axes accounted for 93.3% of the bio-clogging changes, suggesting that the surface properties indeed considerably influenced the bio-clogging characteristics. The roughness (57.6%) could explain the differences in the bio-clogging factors (Table S8). Moreover, a small angle was observed between the roughness and cell copies, suggesting a higher correlation in phase I. As illustrated in Fig. S4a, the bacterial 16 S rRNA gene of the HDPE was dominant ($5.03\pm0.94\times10^5$ copies/cm^2) among different pipe materials, consistent with the highest roughness of the HDPE (Fig. 2). The two axes also indicated a lower variability in phase II (78.4%) than that of phase I (93.3%). The predominant factors were the electron donator parameter (γ^-) and electron acceptor parameter (γ^+) (Table S9). A strong correlation between γ^- and protein was observed, supported by the fact the PVC had the highest γ^- (2.2 mJ/m^2) and protein content (2623.1 ± 33.2 μg/cm^2). These results suggested that the predominant surface properties varied with phases. In the initial period, bio-clogging occurred by the adhesion of bacteria with an increase in the roughness. In the later period, the bio-clogging generation became highly complex with the secretion of proteins and polysaccharides owing to the increase in γ^-.

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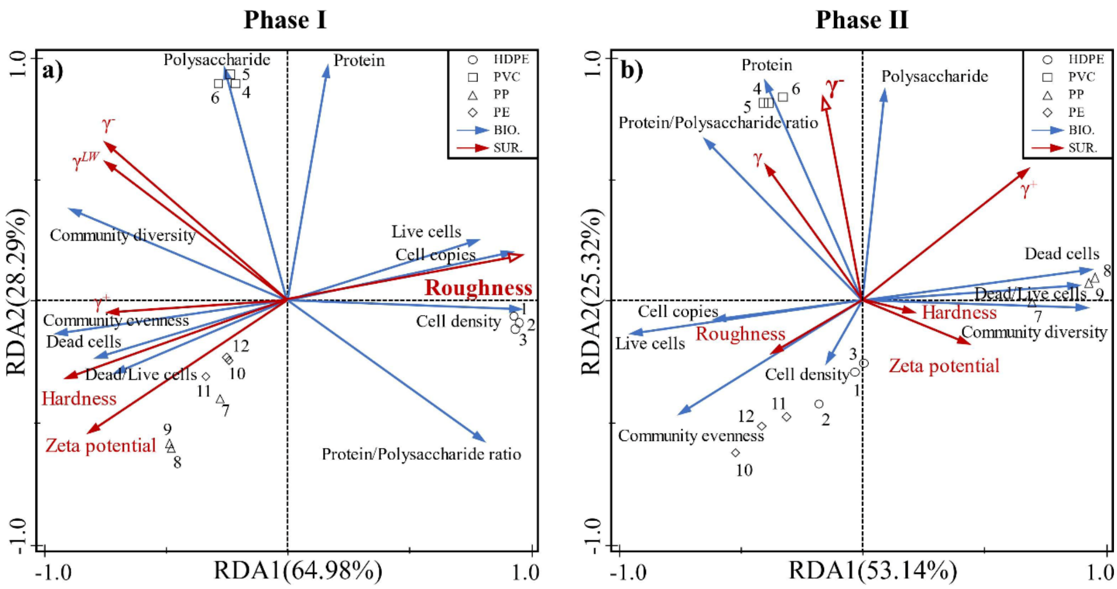

Fig. 7 Redundancy analysis (RDA) of the relationships between the bio-clogging characteristics and physicochemical properties of pipe materials in two different phases.

Moreover, linear correlation tests were performed to investigate the relationship between the bio-clogging characteristics and pipe material physicochemical properties (Fig. 8). The total surface free energy (γ) was significantly correlated with the Lifshitz-van der Waals component (γ^{LW}) (R^2=1.0, P≤0.001), consistent with the fact that γ^{LW} was the main contributor to the γ value of different pipe materials (Fig. 5b). Interestingly, the Zeta potential was positively correlated with the hardness (R^2=0.94, P≤ 0.001). However, limited information is available regarding the relationship between the zeta potential and hardness. In phase I, both the cell density (R^2=0.93) and cell copies (R^2=0.94) were strongly correlated with the roughness (P≤0.001). These results were consistent with the RDA results, indicating that roughness was the predominant factor for bacterial adhesion and promoted bio-clogging formation. In phase II, γ^- was positively correlated with the protein (R^2=0.77) and protein/polysaccharide ratio (R^2=0.73), respectively (P≤0.01). Thus, γ^- could stimulate

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the secretion of proteins and promote bio-clogging formation. In practice, the leachate quality may vary from landfill to landfill, leading to different bio-clogging characteristics. Additional research must be performed on various leachate characteristics to validate the applicability of this study. The research protocols and statistical analysis presented in this study can provide a valuable basis for future studies.

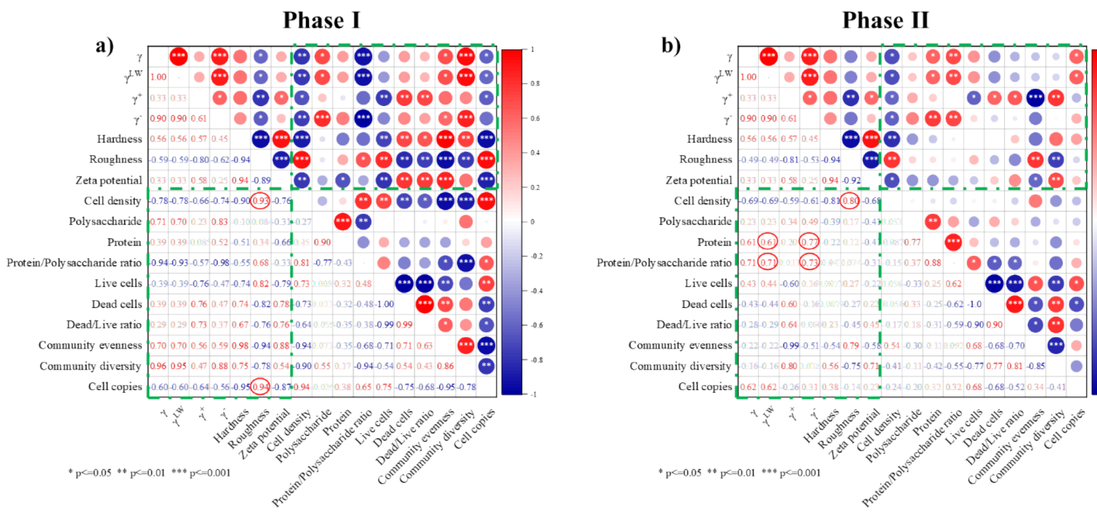

Fig. 8 Person's linear correlation between the surface properties and bio-clogging characteristics.

4. Conclusions

The influence and mechanisms of surface physicochemical properties on bio-clogging formation processes were comprehensively investigated. The bio-clogging characteristics varied with the pipe materials owing to the different surface physicochemical properties. The roughness was the main contributor to the bacterial adhesion and promoted bio-clogging in phase I. The maximum amount of bio-clogging materials was observed in the HDPE, along with the highest roughness (432±76 nm). However, the electron donator parameter (γ^-) was the predominant factor in the bio-clogging development in phase II. The highest γ^- (2.2 mJ/m^2) and protein content

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(2623.1±33.2 μg/cm^2) were observed for the PVC. Moreover, the interaction energy indicated that the bacteria could irreversibly and reversibly adhere to the HDPE, whereas irreversible adhesion occurred on PVC, PP, and PE. This study provides a novel thermodynamic framework for the bio-clogging development in leachate for different pipe materials and can enhance the fundamental understanding of bio-clogging, thereby promoting the development of novel bio-clogging control strategies.

Declaration of competing interest

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

Acknowledgments

This research was financially supported by the National Natural Science Foundation of China (22176005) and Guangdong Basic and Applied Basic Research Foundation (2021A1515110724).

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