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PE-Cd复合污染土壤中Cd释放迁移特征及机制

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摘要: 微塑料广泛分布于土壤环境中, 不仅改变了土壤的理化性质还可富集重金属等污染物, 进而影响它们在环境中的归趋。目前, 有关微塑料对土壤中重金属运移影响的作用机制尚不清晰。选取聚乙烯(PE)微塑料和Cd为研究对象, 在明确PE对Cd²⁺吸附作用机制的基础上, 通过土柱淋溶实验研究了不同离子强度和类型(Ca²⁺、Na⁺)下, PE含量和粒径对土壤中Cd释放迁移行为的影响。结果表明, PE对Cd²⁺的吸附能力随粒径的增大而降低, 以物理吸附为主, 受颗粒内扩散过程控制。PE对Cd释放迁移的影响与CaCl₂浓度有关, 高离子强度下(0.05 mol·L⁻¹和0.1 mol·L⁻¹), PE促进了Cd²⁺的迁移, Cd²⁺的出流浓度分别由6.48 mg·L⁻¹和16.79 mg·L⁻¹升高到7.12 mg·L⁻¹和23.45 mg·L⁻¹; 低离子强度下(0.01 mol·L⁻¹)抑制了Cd的迁移, Cd²⁺的出流浓度由0.66 mg·L⁻¹降低至0.57 mg·L⁻¹; 且PE添加量越大, 促进或抑制作用越显著。同时, Cd的释放迁移也受PE粒径和含量的影响, 当添加量较小(1%、4%)时, 大粒径对Cd²⁺迁移的促进效果更加显著; 而当添加量较大(7%和20%)时, 小粒径更易促进Cd²⁺的迁移。当淋溶液为NaCl时, 土壤的渗透性显著降低, PE对Cd的迁移无显著影响, 但改变了土壤团聚体的稳定性。总之, 微塑料改变了土壤中Cd的释放迁移行为, 作用结果不仅与微塑料的粒径和含量有关, 还受土壤溶液化学性质的影响。

关键词: 聚乙烯(PE); 微塑料; 吸附; 镉(Cd); 淋溶迁移; 土壤

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Characteristics and Mechanism of Cd Release and Transport in Soil Contaminated with PE-Cd

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Abstract: Microplastics (MPs) are a type of emerging contaminants that pose a potential threat to global terrestrial ecosystems. The accumulation of MPs in soil inevitably affects soil physical and chemical properties, both directly and indirectly. Additionally, owing to their small size and surface features, MPs have excellent sorption capacity for both organic and inorganic materials, thus affecting their fate in the environment. However, the influence of MPs on heavy metal sorption and transport in soil is still not fully understood. In this study, polyethylene (PE) and Cd were selected as research objects, and on the basis of clarifying the adsorption mechanism of Cd on PE MPs, the effects of PE concentration and particle size on Cd release and transport behavior in soil under different ionic strengths and types (Ca²⁺ and Na⁺) were studied using column leaching experiments. The results of the batch experiments showed that the adsorption capacity of PE MPs for Cd²⁺ decreased with the increase in particle size. Scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), and Zeta potential were used to analyze the properties of PE MPs and adsorption behavior of Cd²⁺ onto MPs. The adsorption was mainly a physical process and was controlled by intra-particle diffusion. The adsorption kinetics could be described well by the quasi-second-order kinetics and Webber-Morris model. The adsorption isotherm conformed to the Langmuir model, indicating monolayer adsorption. The results of leaching experiments showed that the effect of PE MPs on Cd release and transport in soil was related to the CaCl₂ concentration. At high ionic strength (0.05 mol·L⁻¹ and 0.1 mol·L⁻¹), PE promoted the transport of Cd. The effluent concentration of Cd²⁺ increased from 6.48 mg·L⁻¹ and 16.79 mg·L⁻¹ to 7.12 mg·L⁻¹ and 23.45 mg·L⁻¹, whereas at low ionic strength (0.01 mol·L⁻¹), Cd transport was inhibited by PE MPs, and the effluent concentration of Cd²⁺ decreased from 0.66 mg·L⁻¹ to 0.57 mg·L⁻¹. The larger the amount of PE added, the more significant the promoting or inhibiting effect. Additionally, the release and transport of Cd in soil were also affected by the MPs particle size and concentration. When the addition amount was small (1%, 4%), the large-sized MPs were more conducive to the transport of Cd in soil. When the addition amount was large (7%, 20%), MPs with small particle sizes promoted Cd²⁺ transport more significantly. When the leaching solution used was NaCl, soil permeability decreased significantly. PE MPs had no significant effect on Cd release and transport but changed the stability of soil aggregates. In conclusion, PE MPs could change the release and transport behavior of Cd in soil, and the impact results were not only related to the particle size and content of MPs but were also influenced by the chemical properties of the soil solution.

Key words: polyethylene (PE); microplastics; adsorption; cadmium (Cd); leaching; soil

塑料和塑料制品已被广泛应用于工业领域和人类的日常生活中, 惊人数量的塑料废弃物被释放到陆地、淡水和海洋环境中。进入环境的塑料在风化和降解等外界作用下逐步破碎化而形成一类新兴污染物——微塑料^[1, 2]。微塑料通常指直径小于5 mm的塑料微粒, 已成为日益受到关注的全球性环境问题^[3, 4]。目前, 微塑料已经在世界各地的水体、土壤、沉积物和生物体等基质中被检测到^[5, 6], 其中土壤中

微塑料的丰度约为海洋的4~23倍^[7], 是微塑料重要的汇集地^[8]。对中国5个省份6种不同土地利用类型

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土壤微塑料调查发现,微塑料广泛存在,含量范围为 $2\,783 \sim 6\,366 \text{ items} \cdot \text{kg}^{-1}$ [9]。微塑料主要通过施肥、地膜破损、污水灌溉、垃圾填埋、大气沉降及地表径流等方式进入土壤 [10-12]。进入土壤的微塑料会对土壤理化性质、土壤功能及生物多样性产生显著影响 [13],还可与重金属、抗生素和有机氯农药等污染物结合产生复合污染效应 [14,15],最终对人体健康和生态系统造成潜在威胁。

微塑料与各种污染物的相互作用是一个复杂的过程,受微塑料、污染物及土壤理化性质的影响。微塑料的粒径大小、比表面积及表面特征对污染物的吸附性能起着关键的作用 [16,17]。Zhou 等 [18] 研究表明聚酰胺(PA)表面官能团(如 C—O 和 N—H)的存在会优先与金属离子相互作用,使得 PA 对 Cd^{2+} 的吸附能力更强;同种微塑料对不同重金属的吸附能力也存在较大差异,Wang 等 [19] 研究发现聚对苯二甲酸乙二醇酯(PET)对 Cu 和 Zn 的最大吸附量分别为 $40.3 \mu\text{g} \cdot \text{g}^{-1}$ 和 $24.7 \mu\text{g} \cdot \text{g}^{-1}$ 。另外,微塑料可以改变土壤对污染物的吸附能力。Hüffer 等 [20] 研究表明 PE 微塑料通过竞争、阻挡和稀释等作用降低了土壤对疏水性有机污染物的吸附能力而促进有机污染物的迁移;而赵雪君等 [21] 研究发现 PA 和聚甲基丙烯酸甲酯(PMMA)增强了土壤对农药异菌脲的吸附能力,抑制了异菌脲在土壤中的迁移,且使其不易被降解,从而加剧表层土壤的污染。微塑料还可以通过改变土壤的理化性质,进而影响到土壤中物质的迁移转化。Wen 等 [22] 研究发现微塑料显著改变了黄棕壤的化学性质(pH、OM 和 CEC),进而改变了土壤中重金属的化学形态;Yu 等 [23] 研究表明微塑料可能通过物理吸附或者共沉淀来降低团聚体中重金属的含量,或通过改

变土壤的理化性质来影响不同重金属形态的迁移转化,从而降低重金属的生物利用度。

长期以来重金属被认为是一种典型的土壤污染物,受到世界各国的普遍关注。我国耕地土壤重金属点位超标率达 19.4% [24],其中 Cd 的点位超标率为 7%,已被确定为我国土壤的首要污染物。微塑料的存在会改变重金属在土壤中的迁移、转化及归趋,进而影响其生态毒性。目前,更多的研究集中在水环境中重金属在各类型微塑料上的吸附能力和机制,尚缺乏微塑料与重金属复合污染土壤中重金属迁移特征的研究。因此,本文选取 CaCl_2 、NaCl 为淋洗溶液,通过土柱淋溶实验研究不同离子强度和类型下,聚乙烯(PE)微塑料含量、粒径对土壤中 Cd 吸附迁移行为的影响,以期对土壤中微塑料与重金属复合污染风险评价和防治提供理论依据。

1 材料与方法

1.1 供试材料

聚乙烯(PE)是日常生活及农业生产中最常见的塑料之一,购自东莞市樟木头华创塑胶原料商行,选取 3 种不同粒径: $50 \mu\text{m}$ 、 $500 \mu\text{m}$ 和 1 mm 。将 PE 微塑料置于 5% 的硝酸中浸泡 24 h,去除表面杂质,然后用去离子水洗至中性后低温烘干备用。采用扫描电镜(SEM)和傅里叶红外光谱仪(FTIR)对微塑料的微观形貌和表面基团进行分析。本实验所用土壤采集于青岛市即墨蓝村镇($120^\circ 17' \text{E}$, $36^\circ 42' \text{N}$),土壤类型为棕壤。采集 0~20 cm 表层土壤,去除植物的根、砂石及其他杂物,风干、碾细,过 10 目筛,备用。土壤基本理化性质如表 1 所示。采用纳米粒度 Zeta 电位测定仪测定微塑料及土壤混合样品的 Zeta 电位。

表 1 供试土壤基本理化性质

Table 1 Physical and chemical properties of the tested soil

土壤类型	pH (H_2O)	ω (有机质) / $\text{g} \cdot \text{kg}^{-1}$	阳离子交换量 / $\text{cmol} \cdot \text{kg}^{-1}$	占比/%		
				2~0.05 mm 粒径	0.05~0.002 mm 粒径	<0.002 mm 粒径
棕壤	5.96	22.80	17.98	34.76	42.64	22.6

1.2 污染土壤的制备

参考我国《土壤环境质量 农用地土壤污染风险管控标准(试行)》(GB 15618-2018)及山东省农田实际污染状况 [25],设定土壤 Cd 污染水平为 $20 \text{ mg} \cdot \text{kg}^{-1}$ 。将浓度为 $500 \text{ mg} \cdot \text{L}^{-1}$ 的 $\text{Cd}(\text{NO}_3)_2$ 溶液均匀地喷洒在土壤表面,经充分混合后静置 24 h。然后通过分批均匀铺洒混合的方式向土壤中添加微塑料,为探究极限微塑料添加水平下对土壤中重金属迁移的影响,设置添加比例分别为 0%、1%、4%、7% 和 20% (以微塑料/土壤干重计),依次编号为 CK、T1、T2、T3 和 T4。最后,将 Cd 与 PE 复合污染的土样置于温室中老

化培养,14 d 后进行土柱淋溶实验。

1.3 PE 微塑料对 Cd 的吸附实验

1.3.1 吸附动力学实验

分别取 0.04 g 不同粒径的 PE 微塑料放入离心管中,然后加入 40 mL pH 值为 6.0,浓度为 $6 \text{ mg} \cdot \text{L}^{-1}$ 的 $\text{Cd}(\text{NO}_3)_2$ 溶液,于 25°C 、转速为 $160 \text{ r} \cdot \text{min}^{-1}$ 条件下振荡 0.25、0.5、1、2、5、8、10、12、24 和 48 h。取上清液,过 $0.45 \mu\text{m}$ 滤膜,采用电感耦合等离子体发射光谱仪(ICP-OES)测定滤液中 Cd^{2+} 浓度。每个处理设 3 个重复。微塑料对 Cd^{2+} 的吸附量(Q_t)由以下方程式计算:

$$Q_t = \frac{(c_0 - c_t)V}{m} \quad (1)$$

式中, Q_t 为 PE 微塑料对重金属的吸附量 ($\text{mg} \cdot \text{g}^{-1}$); c_0 为重金属的初始浓度 ($\text{mg} \cdot \text{L}^{-1}$); c_t 为 t 时刻溶液中重金属离子的浓度 ($\text{mg} \cdot \text{L}^{-1}$); V 为加入重金属溶液的体积 (L); m 为加入微塑料的质量 (g).

选用准一级动力学、准二级动力学和颗粒内扩散模型 (Webber-Morris) 对实验数据进行拟合, 表达式如下.

准一级动力学模型:

$$Q_t = Q_e(1 - e^{-k_1 t}) \quad (2)$$

准二级动力学模型:

$$Q_t = \frac{k_2 t Q_e^2}{1 + k_2 t Q_e} \quad (3)$$

Webber-Morris 模型:

$$Q_t = k_i t^{0.5} + C \quad (4)$$

式中, Q_e 为吸附平衡时微塑料的吸附量 ($\text{mg} \cdot \text{g}^{-1}$); k_1 为准一级动力学反应速率常数 (h^{-1}); k_2 为准二级动力学吸附反应速率常数 [$\text{g} \cdot (\text{mg} \cdot \text{h})^{-1}$]; k_i 为颗粒内扩散模型的速率常数 [$\text{mg} \cdot (\text{g} \cdot \text{h}^{1/2})^{-1}$]; C 为与边界层厚度有关的常数.

1.3.2 等温吸附实验

配制浓度为 2、4、6、8、10 和 15 $\text{mg} \cdot \text{L}^{-1}$ 的 $\text{Cd}(\text{NO}_3)_2$ 溶液, 调节 pH 值为 6.0, 分别将 0.04 g 不同粒径的微塑料添加到 40 mL 浓度为 2 ~ 15 $\text{mg} \cdot \text{L}^{-1}$ 的 Cd 溶液中, 在 25℃、转速为 160 $\text{r} \cdot \text{min}^{-1}$ 条件下振荡 48 h, 将上清液经 0.45 μm 的滤膜过滤后测定 Cd^{2+} 浓度, 每个处理设置 3 个重复.

选用 Langmuir 和 Freundlich 方程来描述微塑料对重金属的等温吸附, 表达式分别如下.

Langmuir 等温模型:

$$Q_e = \frac{K_L Q_m c_e}{1 + K_L c_e} \quad (5)$$

Freundlich 等温模型:

$$Q_e = K_F c_e^{1/n} \quad (6)$$

式中, Q_m 为微塑料对重金属离子的最大吸附量 ($\text{mg} \cdot \text{g}^{-1}$); K_L 为与吸附相关的常数 ($\text{L} \cdot \text{mg}^{-1}$); c_e 为溶液中重金属离子的平衡浓度 ($\text{mg} \cdot \text{L}^{-1}$); K_F 为 Freundlich 吸附系数 ($\text{mg}^{1-n} \cdot \text{L}^n \cdot \text{g}^{-1}$); $1/n$ 为吸附强度.

1.4 土柱淋溶实验

根据实测土壤容重 1.30 $\text{g} \cdot \text{cm}^{-3}$ 填装土柱 (容器为有机玻璃, 高度 20 cm, 内径 5 cm). 将培养后的污染土壤分 4 次填装到土柱中, 以保证土壤颗粒均匀分布; 不同微塑料处理的土柱中, 土壤质量相同. 在土柱两端各铺一层石英砂, 以保证供液时溶液能均匀地入渗到土壤中, 避免土壤颗粒堵塞取样口. 用蠕动

泵作为供水装置自下而上通入去离子水对土柱进行饱和 (排出柱内空气), 待饱和后调换入流方向. 水流稳定后 (1 $\text{mL} \cdot \text{min}^{-1}$), 通入 $\text{CaCl}_2/\text{NaCl}$ 溶液进行淋溶, 6 h 后改用去离子水淋洗至无 Cd^{2+} 流出. 用自动部分收集器对流出液连续取样, 测定 pH 及 Cd^{2+} 浓度. 具体实验设置见表 2.

2 结果与讨论

2.1 PE 微塑料对 Cd 的吸附特性

2.1.1 样品的表征

图 1(a) ~ 1(c) 分别为 50 μm 、500 μm 及 1 mm 的 PE 微塑料扫描电镜 (SEM) 结果, 可以观察到粒径为 50 μm 的 PE 微塑料颗粒表面比较光滑平整且存在大量的孔隙结构; 粒径为 500 μm 和 1 mm 的微塑料呈不规则几何形, 表面较粗糙且有明显的褶皱和凸起. EDS 分析发现 [图 1(d)], 吸附 Cd 后的 PE 微塑料表面检测到 Cd 的存在, 且分布均匀, 说明微塑料表面对 Cd^{2+} 的吸附点位较为均一, 以单层吸附为主. 由于微塑料表面含氧官能团的脱质子化作用^[26], 不同溶液化学条件下微塑料表面带有负电荷 (-2.16 ~ -6.25 mV), 但数量较少, 少量 Cd^{2+} 可通过静电吸引被微塑料吸附.

如图 2 所示, PE 微塑料的特征峰可以表述在 2 914 cm^{-1} 处— CH_2 不对称伸缩振动、2 847 cm^{-1} 处— CH_2 对称伸缩振动、1 472 cm^{-1} 处— CH_2 弯曲振动和 721 ~ 717 cm^{-1} 处— CH_2 面内摇摆振动^[27]. 这些官能团对 Cd 无显著吸附作用^[28], 故物理作用在吸附过程中占主导地位.

2.1.2 吸附动力学特征

如图 3 所示, PE 对 Cd^{2+} 的吸附过程主要分为 3 个阶段: 第一阶段为快速吸附阶段 (0 ~ 1 h), 由于吸附开始时微塑料表面吸附点位充足, Cd^{2+} 吸附速率较快, 约 67% 的 Cd^{2+} 被吸附; 第二阶段为慢速吸附阶段 (1 ~ 10 h), 随着吸附反应的进行, 微塑料上的吸附点位逐渐被占满, 微塑料与溶液中 Cd^{2+} 的浓度差降低, Cd^{2+} 进入微塑料表面的褶皱微孔, 吸附量增长变缓; 第三阶段为平衡吸附阶段 (10 ~ 48 h), 此时吸附逐渐达到平衡, 吸附量变化很小. 3 种粒径中 50 μm 微塑料因其比表面积最大, 提供的点位较多, 所以吸附容量最大. 采用准一级动力学模型和准二级动力学模型对 Cd^{2+} 的吸附过程进行拟合, 结果如表 3 所示. 准二级动力学模型的 R^2 值 (0.921 ~ 0.970) 均高于准一级动力学模型 (0.737 ~ 0.901), 且拟合获得的 Q_e 更接近实测值 (0.948、1.007 和 1.129 $\text{mg} \cdot \text{g}^{-1}$), 表明准二级动力学模型能更好地描述 PE 对 Cd^{2+} 的吸附过程, 吸附过程受表面吸附位点、传质和颗粒内扩散等作用影响.

为了进一步研究 PE 对 Cd^{2+} 的吸附过程, 采用

表 2 实验设置¹⁾
Table 2 Experimental conditions

柱序号	土壤质量/g	污染土壤编号	PE 添加量(质量分数)/%	PE 粒径	离子强度/mol·L ⁻¹	离子类型
1	281	CK	0	—	0.01	CaCl ₂
2	281	CK	0	—	0.05	CaCl ₂
3	281	CK	0	—	0.1	CaCl ₂
4	281	CK	0	—	0.1	NaCl
5	281	T1	1	50 μm	0.01	CaCl ₂
6	281	T1	1	50 μm	0.05	CaCl ₂
7	281	T1	1	50 μm	0.1	CaCl ₂
8	281	T1	1	50 μm	0.1	NaCl
9	281	T1	1	500 μm	0.1	CaCl ₂
10	281	T1	1	1 mm	0.1	CaCl ₂
11	281	T2	4	50 μm	0.01	CaCl ₂
12	281	T2	4	50 μm	0.05	CaCl ₂
13	281	T2	4	50 μm	0.1	CaCl ₂
14	281	T2	4	50 μm	0.1	NaCl
15	281	T2	4	500 μm	0.1	CaCl ₂
16	281	T2	4	1 mm	0.1	CaCl ₂
17	281	T3	7	50 μm	0.01	CaCl ₂
18	281	T3	7	50 μm	0.05	CaCl ₂
19	281	T3	7	50 μm	0.1	CaCl ₂
20	281	T3	7	50 μm	0.1	NaCl
21	281	T3	7	500 μm	0.1	CaCl ₂
22	281	T3	7	1 mm	0.1	CaCl ₂
23	281	T4	20	50 μm	0.01	CaCl ₂
24	281	T4	20	50 μm	0.05	CaCl ₂
25	281	T4	20	50 μm	0.1	CaCl ₂
26	281	T4	20	50 μm	0.1	NaCl
27	281	T4	20	500 μm	0.1	CaCl ₂
28	281	T4	20	1 mm	0.1	CaCl ₂

1)“—”表示无微塑料添加

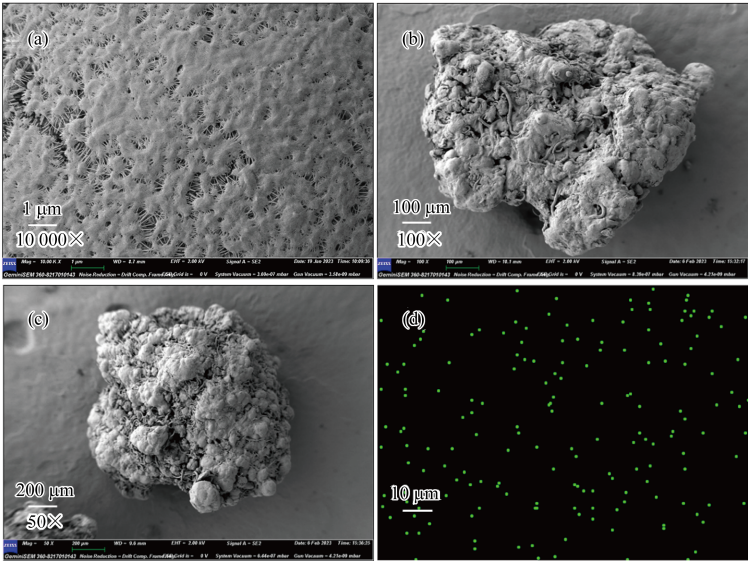


图 1 PE 微塑料的 SEM 扫描图及 EDS 能谱图
Fig. 1 SEM scanning and EDS spectra of PE microplastics

Webber-Morris 模型对吸附结果进行拟合,如图 3 和表 4 所示. 吸附过程分为 3 个阶段,分别为液膜扩散、颗粒内扩散和吸附/解吸动态平衡阶段. 3 个阶段的扩

散速率常数大小排序为: $k_1 > k_2 > k_3$, 且 C 值不等于 0, 表明微塑料对 Cd^{2+} 的吸附速率不仅受颗粒内扩散控制, 外部扩散也很重要^[29].

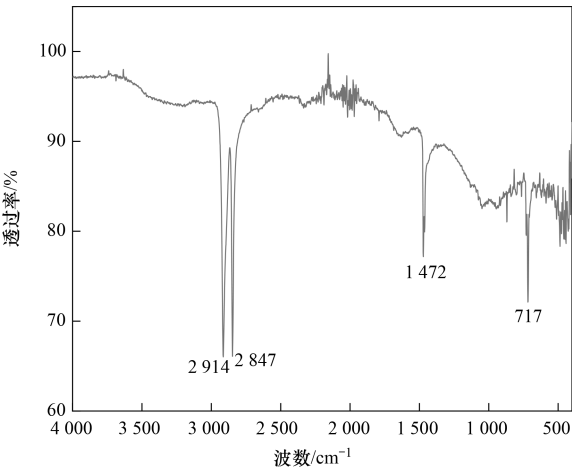


图2 PE微塑料的FTIR谱图

Fig. 2 FTIR spectra of PE microplastics

2.1.3 吸附等温线

图4为不同粒径PE微塑料对Cd²⁺的吸附等温线,随着平衡液浓度的增加,PE对Cd²⁺的吸附量也逐渐增加.当Cd²⁺浓度达15 mg·L⁻¹时,3种粒径(50 μm、500 μm和1 mm)的微塑料对应的吸附量分别为1.620、1.435和1.320 mg·g⁻¹,微塑料粒径越小,吸附

量越大.采用Langmuir和Freundlich模型对Cd²⁺的等温吸附进行拟合(表5),Langmuir模型的拟合结果(R^2 值在0.989~0.995之间)优于Freundlich模型(R^2 值在0.917~0.963之间),表明PE对Cd²⁺的吸附是单分子层吸附^[30],微孔填充是微塑料吸附Cd²⁺的主要机制. Q_m 随粒径的减小而增大,与本实验的结果一致,说明Cd²⁺在塑料颗粒上的吸附与其尺寸大小密切相关,这是因为较小的塑料颗粒具有较大的比表面积,因此增加了固液接触的机会,从而为Cd²⁺提供了更多的吸附位点^[31]. K_L 可用来反映微塑料对Cd²⁺的吸附强度, K_L 值越大自发反应速率也越大,从表中可以看出,随着微塑料粒径的减小 K_L 增大.此外, $1/n$ 可用于评估吸附类型,当 $1/n$ 大于1时,吸附是化学吸附,当 $1/n$ 小于1时,是物理吸附^[32].如表5所示, $1/n$ (0.381~0.462)的值均小于1,进一步表明微塑料对Cd²⁺的吸附以物理吸附为主.

2.2 PE微塑料添加量对土壤中Cd²⁺释放迁移的影响

通过柱实验研究了不同微塑料处理下Cd在土壤中的迁移特征.图5为0.1 mol·L⁻¹ CaCl₂条件下,不同

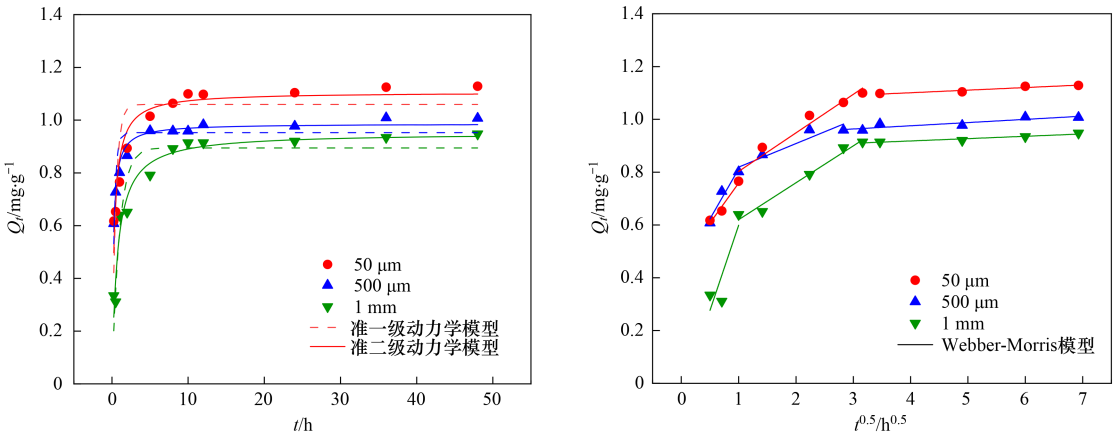


图3 PE微塑料吸附Cd²⁺的动力学曲线及模型拟合

Fig. 3 Adsorption kinetics of Cd²⁺ fitted with kinetic models

表3 PE微塑料吸附Cd²⁺的动力学模型拟合参数

Table 3 Fitted parameters of the kinetic model of Cd²⁺ adsorption on PE microplastics

PE 粒径	准一级动力学模型			准二级动力学模型		
	$Q_e/\text{mg}\cdot\text{g}^{-1}$	k_1/h^{-1}	R^2	$Q_e/\text{mg}\cdot\text{g}^{-1}$	$k_2/\text{g}\cdot(\text{mg}\cdot\text{h})^{-1}$	R^2
50μm	1.060	2.027	0.737	1.106	2.954	0.921
500μm	0.946	3.336	0.804	0.978	5.991	0.970
1mm	0.895	1.020	0.901	0.952	1.523	0.953

表4 PE微塑料吸附Cd²⁺的Weber-Morris模型拟合参数

Table 4 Fitted parameters of the Weber-Morris model of Cd²⁺ adsorption on PE microplastics

PE 粒径	阶段一			阶段二			阶段三		
	C_1	k_1	R_1^2	C_2	k_2	R_2^2	C_3	k_3	R_3^2
50 μm	0.456	0.303	0.963	0.657	0.146	0.951	1.061	0.009 8	0.918
500 μm	0.434	0.378	0.947	0.730	0.089	0.894	0.927	0.012 0	0.831
1 mm	-0.046	0.645	0.780	0.480	0.140	0.978	0.883	0.008 8	0.925

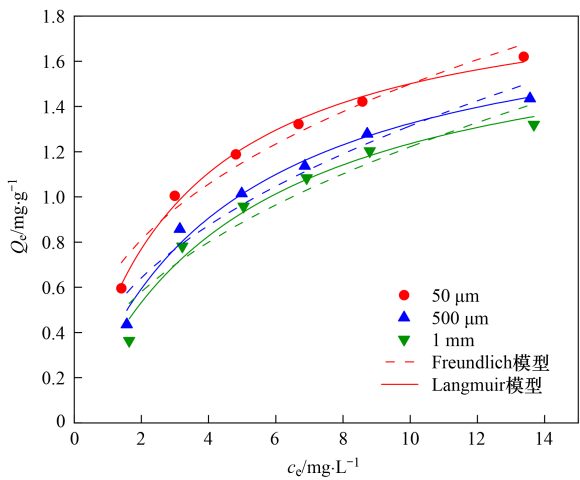


图4 Cd²⁺在PE微塑料上的吸附等温线

Fig. 4 Adsorption isotherms of Cd²⁺ on PE microplastics

微塑料添加量土柱中Cd²⁺的穿透曲线. 淋出液中Cd²⁺浓度峰值随PE添加量的增大而增大,不同处理下(CK、T1、T2、T3和T4), $\rho(\text{Cd}^{2+})$ 峰值分别为16.79、17.66、18.75、20.54和23.45 mg·L⁻¹,且在T2、T3和T4处理下出现峰值提前现象,说明PE微塑料使Cd在土壤中的移动性增强,可导致Cd的生物有效性升高.微塑料的添加为Cd²⁺提供了吸附位点,降低了土

壤对Cd²⁺的吸附,由2.1节的分析可知,微塑料对Cd²⁺的吸附以物理吸附为主,容易通过离子交换、解离等作用释放出一定含量的Cd²⁺[33],从而增加了Cd²⁺的移动性.此外,当微塑料进入土壤后,微塑料与土壤颗粒相结合,使土壤的电位增加(-4.26 mV),进而降低了其对重金属的静电吸附[34],促进了Cd²⁺的迁移.微塑料的添加量越大,对Cd释放迁移的促进作用越强.不同微塑料处理下,Cd²⁺穿透曲线均出现了拖尾现象,说明土壤/微塑料对Cd的吸附解吸受时间控制,这与微塑料对Cd²⁺的动力学吸附实验结果一致.

从图5可以看出,随着CaCl₂溶液的通入,pH呈现下降、趋于平稳、上升和趋于平稳的变化规律.这是因为大量Ca²⁺的输入把Cd²⁺置换出来的同时把土壤上的H⁺置换出来,造成出流液pH降低,有利于Cd²⁺的进一步解吸;去离子水淋洗后,由于土壤的缓冲性及Ca²⁺的解吸,溶液中的H⁺又被吸附,pH逐渐升高,趋于平稳.实验过程中,pH<8,Cd主要以Cd²⁺形式存在,无Cd沉淀产生[35].另外,不同处理土壤pH变化差异不显著,说明PE微塑料对土壤pH影响不大,实验条件下微塑料表面含氧官能团去质子化作用较弱.

表5 PE微塑料吸附Cd²⁺的Langmuir和Freundlich拟合参数

Table 5 Fitted parameters of different isotherm models of Cd²⁺ adsorption on PE microplastics

PE 粒径	Langmuir 模型			Freundlich 模型		
	$Q_m / \text{mg} \cdot \text{g}^{-1}$	$K_L / \text{L} \cdot \text{mg}^{-1}$	R^2	$K_F / \text{mg}^{-n} \cdot \text{L}^n \cdot \text{g}^{-1}$	$1/n$	R^2
50 μm	2.003	0.319	0.995	0.623	0.381	0.963
500 μm	1.685	0.222	0.989	0.472	0.445	0.945
1 mm	1.442	0.172	0.993	0.421	0.462	0.917

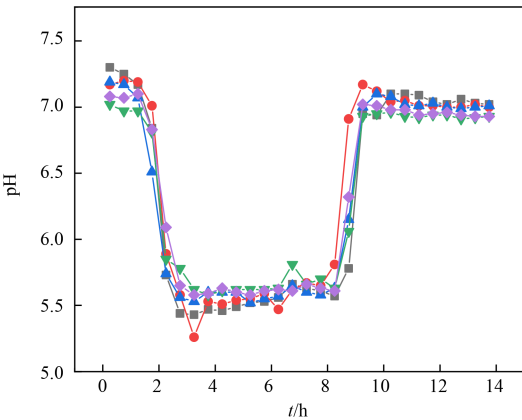
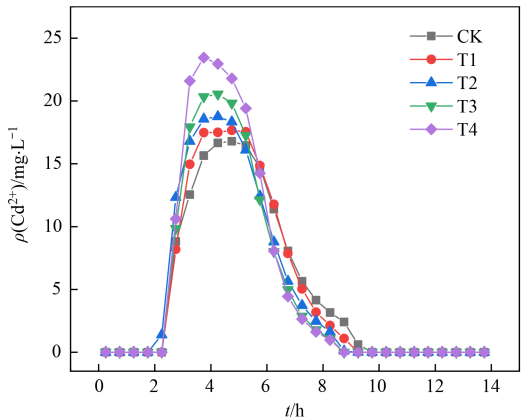


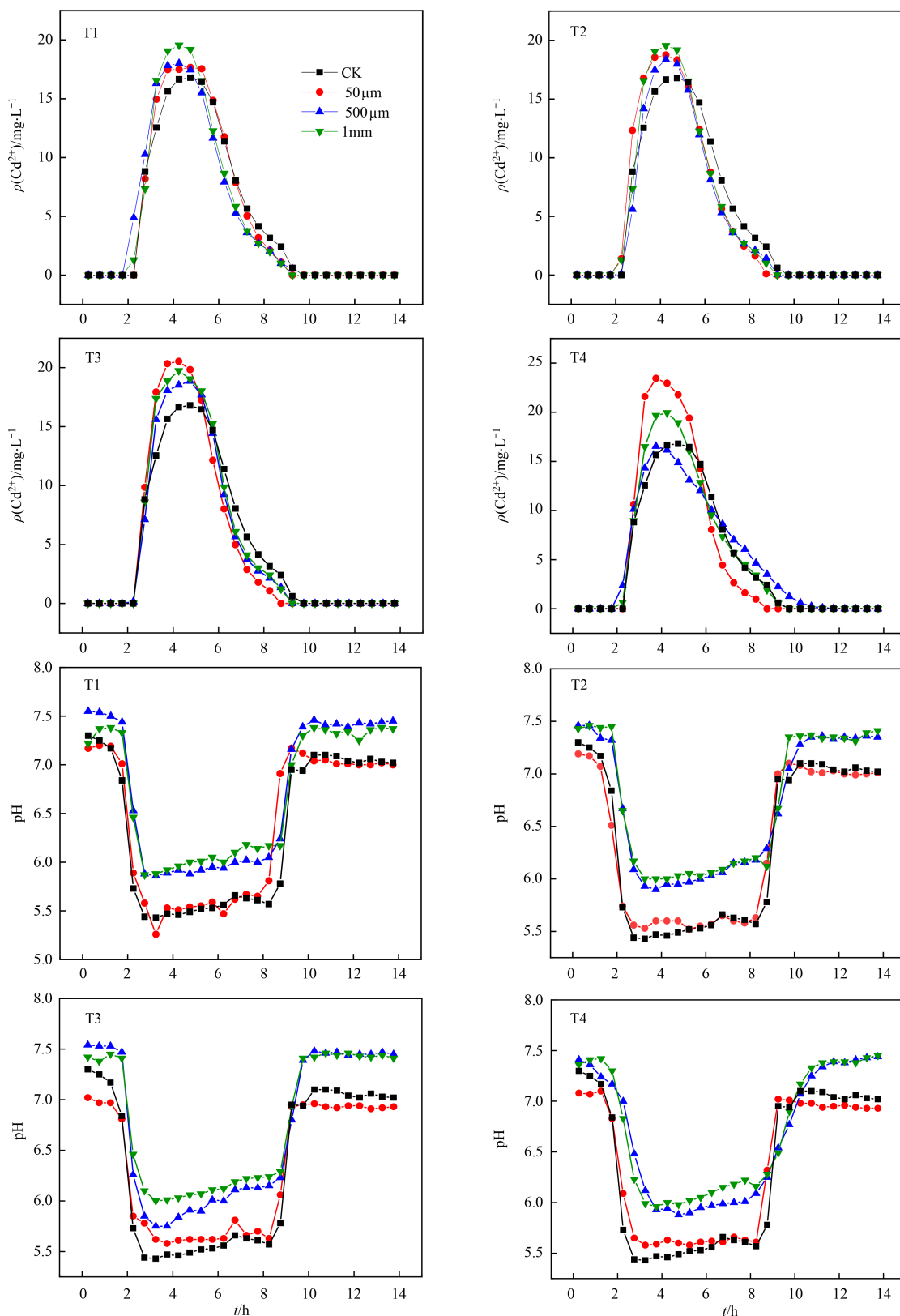
图5 不同PE微塑料添加量下Cd²⁺浓度和pH变化曲线

Fig. 5 Variation of Cd²⁺ concentration and pH under different PE additions

2.3 PE微塑料粒径对Cd²⁺释放迁移的影响

图6为不同粒径(50 μm、500 μm和1 mm)微塑料处理土壤中Cd²⁺淋出浓度变化曲线.从图6中可以看出,除粒径500 μm、添加量20%处理外,PE微塑料均对Cd的迁移有促进作用.当微塑料添加量较大时(7%、20%),小粒径微塑料颗粒(50 μm)更有利于

Cd²⁺的迁移,这是因为微塑料粒径越小,数量也就越多,其与土壤的总接触面积越大,土壤对Cd吸附位点越少;另外,由吸附实验结果可知,PE粒径越小对Cd²⁺的吸附量越大,被PE吸附的Cd²⁺容易解吸,从而促进了Cd²⁺的迁移.微塑料粒径为50 μm时,出流液pH明显低于其他粒径,低pH有利于Cd²⁺的迁移.当

图6 不同PE微塑料粒径下Cd²⁺浓度和pH变化曲线Fig. 6 Variation of Cd²⁺ concentration and pH under different PE particle sizes

微塑料添加量较低时(1%、4%),微塑料对Cd吸附解吸的影响较弱,土壤中大孔隙占比随微塑料粒径的增大而增加^[36],此时土壤中的大孔隙为影响重金属运移的主要因素,从而导致大粒径微塑料(1 mm)对

Cd²⁺迁移的促进作用更显著. de Souza Machado 等^[37]研究发现微塑料颗粒与土壤颗粒相差越大对土壤结构的影响越大,由供试土壤的颗粒组成可以判断1 mm的微塑料对土壤结构的影响最大. 另外,大粒径

微塑料的加入使得微塑料和土壤之间、微塑料和微塑料之间形成新的水流通通道,且微塑料由于本身具有疏水性会使新的水流通通道的水流速度高于原有通道^[38],造成淋出液中 Cd^{2+} 浓度峰值较无微塑料添加土壤提前和增大. 因此,微塑料粒径对 Cd^{2+} 释放迁移的影响与其含量有关,且微塑料含量越大,土壤中 Cd 的移动性对粒径变化越敏感.

2.4 不同 CaCl_2 离子强度下 PE 对 Cd^{2+} 释放迁移的影响

图 7 为 $0.01 \text{ mol} \cdot \text{L}^{-1}$ 和 $0.05 \text{ mol} \cdot \text{L}^{-1}$ CaCl_2 条件下,不同微塑料处理土壤中 Cd^{2+} 淋出浓度变化曲线. 用 CaCl_2 溶液淋溶土柱 2 h 后, Cd^{2+} 浓度逐渐增大,且维持一定水平,去离子水淋溶 2 h 后 Cd^{2+} 浓度逐渐下降. 结合图 5 可以看出,不同处理土壤淋出液中 Cd^{2+} 浓度最大峰值随离子强度的增大而升高,从 $0.66 \text{ mg} \cdot \text{L}^{-1}$ 升高到 $7.05 \text{ mg} \cdot \text{L}^{-1}$ 和 $23.45 \text{ mg} \cdot \text{L}^{-1}$,这是因为 Ca^{2+} 与

Cd^{2+} 竞争土壤/微塑料表面的吸附点位,抑制固相介质对重金属的吸附,从而促进 Cd^{2+} 向下迁移,且 Ca^{2+} 浓度越高,竞争作用越强, Cd^{2+} 淋出浓度越高. H^+ 浓度的变化与 Cd^{2+} 相似, Ca^{2+} 浓度越高,置换出的 H^+ 浓度越高, pH 越低. 在 CaCl_2 淋溶阶段,从释放时间来看,离子强度较大时 ($0.1 \text{ mol} \cdot \text{L}^{-1}$ CaCl_2 , 图 5), Cd^{2+} 在较短时间内被解吸出来, Cd^{2+} 出流浓度在去离子水淋洗前已达到峰值并开始下降;当离子强度较低时 ($0.01 \text{ mol} \cdot \text{L}^{-1}$ 和 $0.05 \text{ mol} \cdot \text{L}^{-1}$ CaCl_2), 通入去离子水后, Cd^{2+} 仍保持较高的释放水平, 2 h 后出流浓度逐渐降低. Cd^{2+} 淋出浓度逐渐降低直至后期不再出流,一方面是因为土壤中大部分 Cd^{2+} 在 CaCl_2 淋溶阶段较容易以可交换态的形式解吸释放出来^[39];另一方面,在去离子水淋溶阶段淋出液 pH 升高,土壤表面的含氧基团得到活化, Cd^{2+} 被络合而不易被解吸出来^[40].

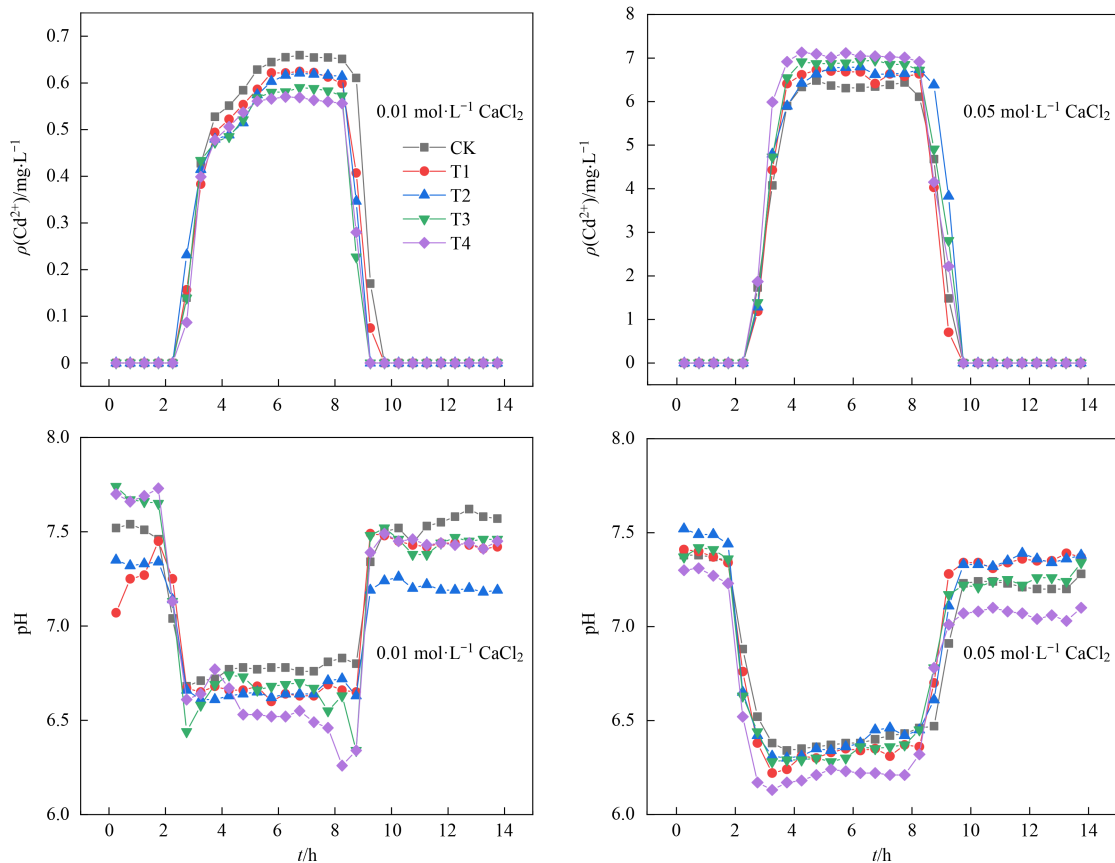


图 7 不同离子强度下 Cd^{2+} 浓度和 pH 变化曲线

Fig. 7 Variation of Cd^{2+} concentration and pH under different ionic strength

不同 CaCl_2 浓度条件下, PE 微塑料对 Cd 迁移的影响不同. 当离子强度较低时 ($0.01 \text{ mol} \cdot \text{L}^{-1}$ CaCl_2), PE 微塑料抑制了 Cd^{2+} 的迁移, 淋出液中 Cd^{2+} 浓度低于未添加微塑料的土壤, 且淋出液中 Cd^{2+} 浓度随 PE 添加量的增加从 $0.66 \text{ mg} \cdot \text{L}^{-1}$ 降低为 $0.57 \text{ mg} \cdot \text{L}^{-1}$. 从图 7 可以看出, 在较低离子强度下淋出液 pH 值较高, 与

金属离子竞争的 H^+ 减少, 微塑料表面官能团易发生脱质子化^[41], 从而导致微塑料表面的电负性 (电位为 -6.25 mV) 和静电吸附位点增加, 使得微塑料对重金属的吸附能力提高, 而且由于 Ca^{2+} 浓度较低, 对 Cd^{2+} 的置换解吸作用较弱, 微塑料对 Cd^{2+} 吸附滞留作用突出, 从而抑制了 Cd^{2+} 的出流. 当离子强度较高时

($0.05 \text{ mol} \cdot \text{L}^{-1}$), 如 2.2 节所示, PE 微塑料促进了 Cd^{2+} 的迁移, 淋出液中 Cd^{2+} 浓度高于未添加微塑料的土壤, 且淋出液中 Cd^{2+} 浓度随微塑料添加量的增加从 $6.48 \text{ mg} \cdot \text{L}^{-1}$ 增大到 $7.12 \text{ mg} \cdot \text{L}^{-1}$. 随着离子强度的增加, 淋出液 pH 值降低, H^+ 与 Ca^{2+} 对微塑料吸附位点的竞争加剧, 溶液中离子化学物质的活性降低以及双电层的压缩^[42], 使得微塑料表面电位升高(图 8), 抑制了微塑料对重金属吸附.

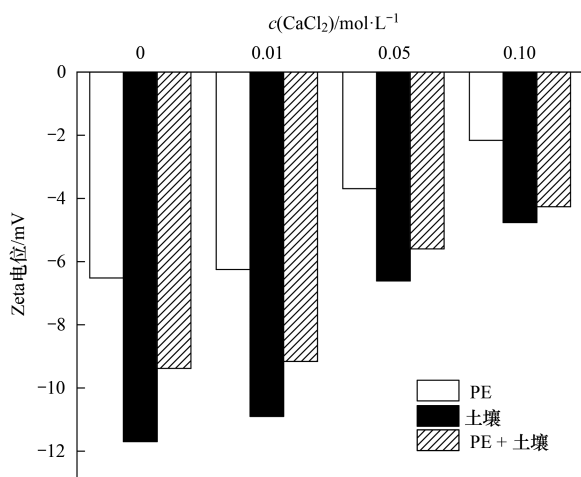


图 8 不同离子强度下微塑料、土壤及其混合物的 Zeta 电位

Fig. 8 Zeta potential of microplastics, soils, and their mixtures at different ionic strengths

2.5 NaCl 条件下 PE 对 Cd^{2+} 释放迁移的影响

Na^+ 为一价阳离子, 与 Ca^{2+} 相比, 对 Cd^{2+} 的竞争吸附及介质表面电位的影响不同. 图 9 为 $0.1 \text{ mol} \cdot \text{L}^{-1}$ NaCl 条件下, 不同微塑料处理土壤中 Cd^{2+} 浓度变化曲线. 总体来看, 由于 Na^+ 的交换选择性系数较低^[43], 交换能力较弱, 出流液中 Cd^{2+} 的浓度较 $0.1 \text{ mol} \cdot \text{L}^{-1}$ CaCl_2 条件下显著下降. 另外, 出流液 pH 高于 $0.1 \text{ mol} \cdot \text{L}^{-1}$ CaCl_2 条件下的 pH 值, 且降低到最低值(约 6.2)后, 急剧升高, 从而抑制了 Cd^{2+} 的迁移. 随着 Na^+ 的不断输入, 不同微塑料处理土壤 Cd^{2+} 淋出浓度均呈现先上升后下降的趋势, 除 T4(20%) 处理外, Cd^{2+} 的穿透曲线基本重合. 在去离子水淋溶阶段, 流速变慢(图 9), 不同处理的土柱中均出现土壤颗粒流出现象. 这是因为可交换性 Na 的含量会极大地影响土壤颗粒分散作用的强弱^[44], 导致土壤团聚体分散成单个土壤颗粒, 通入去离子水后, 小颗粒被活化随溶液流出. 土壤颗粒的分散可由 $0.1 \text{ mol} \cdot \text{L}^{-1}$ NaCl 条件下微塑料和土壤颗粒的表面电位的降低来解释(微塑料、土壤及微塑料与土壤混合的电位分别为 -11.1 、 -21.3 和 -22.0 mV), 电位的降低使颗粒间的静电斥力增强, 有利于颗粒分散. 分散土壤颗粒紧密排列, 使土壤大孔隙数量减小^[45], 出现渗透性差与无法出流的现象. 而 Ca^{2+} 为二价阳离子, 可在土壤颗粒

之间形成桥梁, 即桥键作用, 抑制土壤的分散.

当微塑料无添加或添加量较小时(0%、1%、4% 和 7%), 用去离子水淋洗土柱 2 h, 流速降为 0(图 9); 而 T4(20%) 处理下, 虽有出流但流速逐渐减低, 淋出液中有大量土壤颗粒, 这进一步说明微塑料可改变土壤结构和容重^[46], 大量微塑料的存在使微塑料和土壤之间、微塑料和微塑料之间形成新的水流通路, 土壤颗粒和 Cd^{2+} 可通过这些通道流出; 另外, 微塑料可与土壤颗粒结合, 改变土壤团聚体大小分布, 从而改变孔隙分布, 土壤中微塑料含量越高, 这种影响越显著.

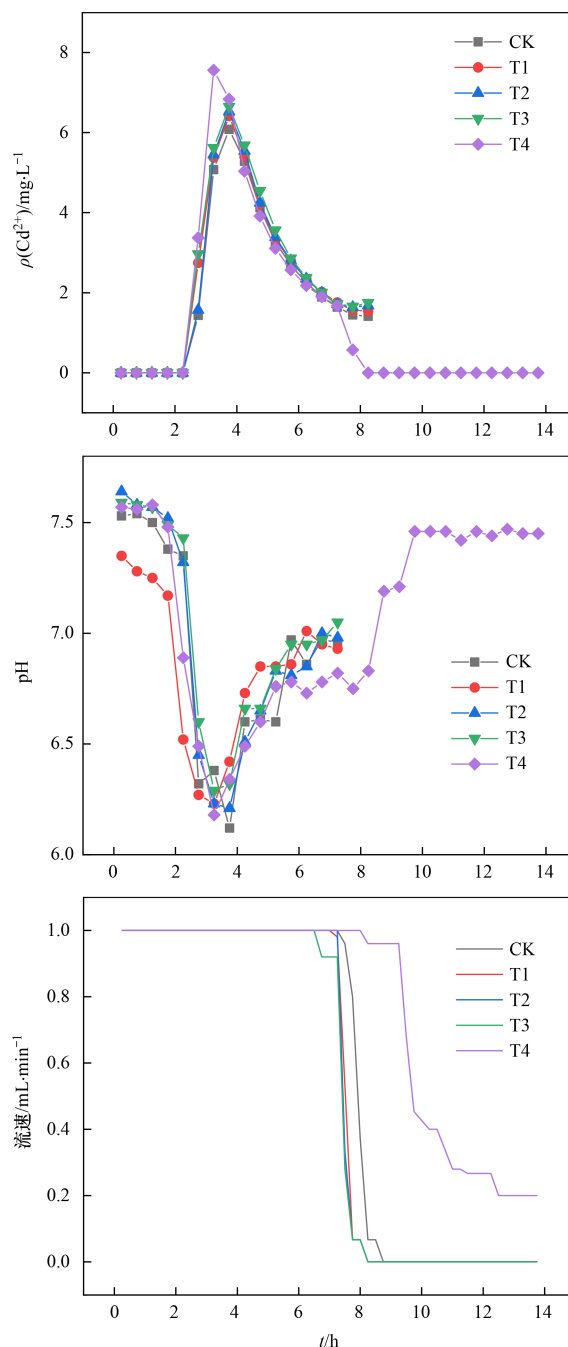


图 9 NaCl 条件下 Cd^{2+} 浓度、pH 值及流速变化曲线

Fig. 9 Variation in Cd^{2+} concentration, pH value, and flow velocity under NaCl condition

3 结论

(1) 微塑料对 Cd^{2+} 的吸附能力随粒径的增大而降低, 以物理吸附为主; 准二级动力学和 Webber-Morris 模型可以很好地拟合 PE 微塑料吸附 Cd^{2+} 的动力学过程, 说明吸附过程是多个吸附阶段共同作用的结果; Langmuir 模型对吸附等温线的拟合效果较好, 证实 PE 微塑料对 Cd^{2+} 的吸附属于单层吸附。

(2) PE 对 Cd 释放迁移的影响与 CaCl_2 浓度有关。高离子强度下 ($0.05 \text{ mol} \cdot \text{L}^{-1}$ 和 $0.1 \text{ mol} \cdot \text{L}^{-1}$), 微塑料使土壤吸附位点减少、电负性降低, 导致土壤对 Cd^{2+} 的吸附能力下降, 且微塑料吸附的 Cd^{2+} 容易发生解吸, 从而促进了 Cd^{2+} 的迁移, 微塑料的添加量越大对 Cd^{2+} 迁移的促进作用越显著; 低离子强度下 ($0.01 \text{ mol} \cdot \text{L}^{-1}$), 由于 Ca^{2+} 的交换作用较弱, 且土壤/微塑料电负性较高, PE 抑制了 Cd^{2+} 的迁移。

(3) PE 对 Cd 释放迁移的影响与微塑料粒径和含量有关。当 PE 添加量较小 (1%、4%) 时, 大粒径微塑料颗粒使得土壤中孔隙所占的比例升高从而更有利于 Cd^{2+} 的迁移; 而当添加量较大 (7%、20%) 时, 微塑料对 Cd^{2+} 吸附解吸的影响增强, 小粒径微塑料与土壤的接触面积较大从而减少土壤中的吸附位点, 更易促进 Cd^{2+} 的迁移。此外, PE 微塑料含量越高, 土壤中 Cd 的移动性对粒径变化越敏感。

(4) NaCl 条件下, Na^+ 显著降低了土壤的渗透性, PE 对 Cd 的迁移无显著影响。 Na^+ 对渗透性的影响与 PE 的添加量有关, 20% 添加量下土壤的渗透性显著增加, 说明 PE 改变了土壤团聚体的稳定性。

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