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Diffusiophoresis: a novel transport mechanism - fundamentals, applications, and future opportunities

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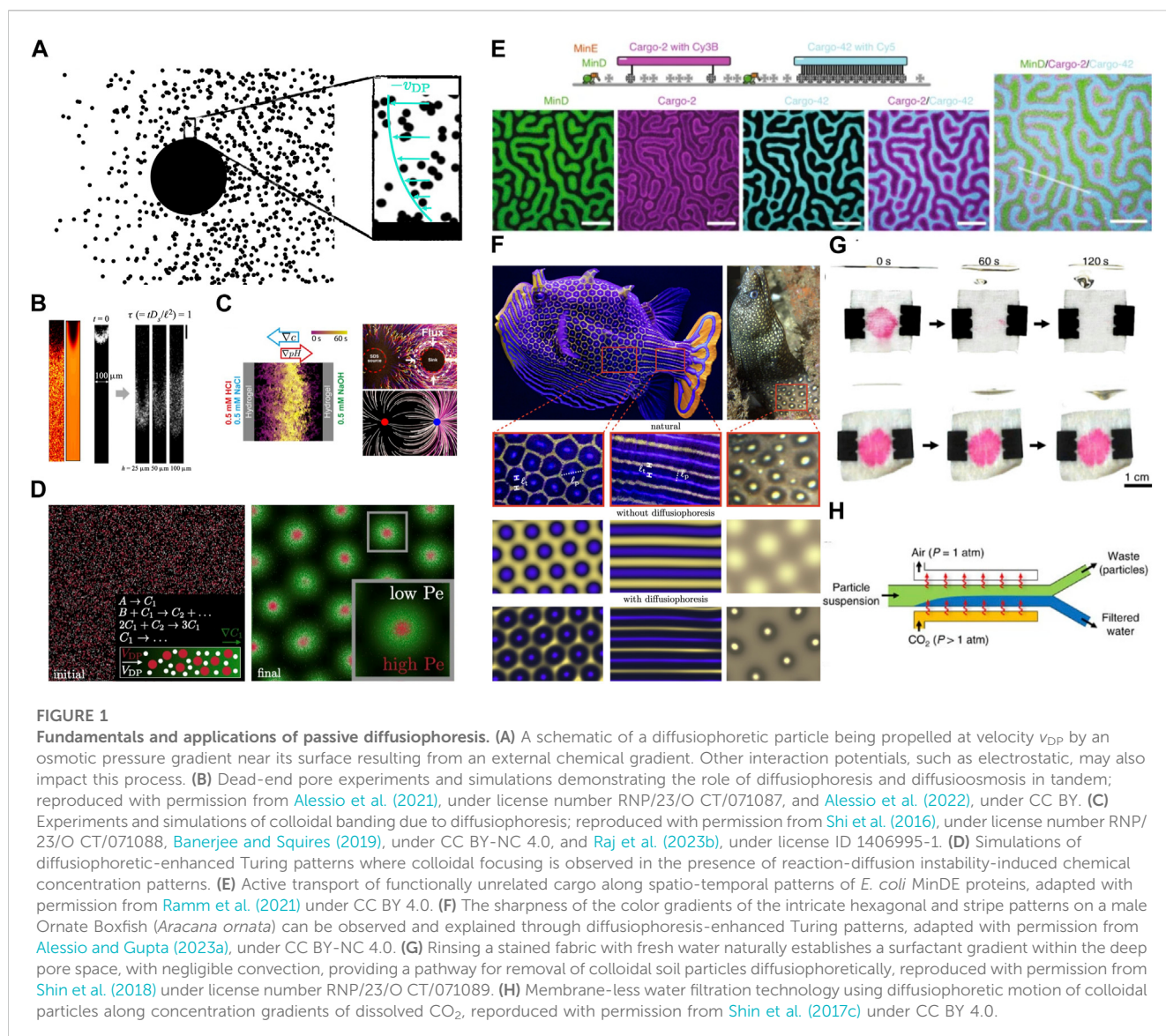
Diffusiophoresis involves the movement of colloidal-scale entities in response to concentration gradients of a solute. It is broadly categorized into two types: passive and active diffusiophoresis. In passive diffusiophoresis, external concentration gradients drive the motion, while in active diffusiophoresis, the colloidal entity itself assists in generating the gradients. In this perspective, we delve into the fundamental processes underlying passive and active diffusiophoresis and emphasize how prevalent both kinds of diffusiophoresis are in colloidal and natural systems. In particular, we highlight the colloidal focusing feature in passive diffusiophoresis and discuss how it underpins the variety of experimental observations and applications such as low-cost zetasizers, water filtration, and biological pattern formation. For active diffusiophoresis, we emphasize the dependence of particle trajectory on its shape and surface heterogeneity, and discuss how this dictates the applications such as drug delivery, removal of microplastics, and self-repairing materials. Finally, we offer insights and ideas regarding future opportunities in diffusiophoresis.

KEYWORDS

colloids, Turing patterns, microfluidics, microbot, propulsion, differential equations, biomedical

1 Introduction

The term ‘diffusiophoresis’ is a combination of the words ‘diffusion’ and ‘phoresis’ and requires two components: a particle and a diffusing species. The diffusing species is typically dissolved in a solvent, and thus, we refer to it as a solute. During diffusiophoresis, the particle moves along the concentration gradients of the solute. Although it may seem counterintuitive, the particle often (though not always) moves towards areas of higher concentration of the solute with which it is interacting. The concept of diffusiophoresis was first documented in 1947 by Derjaguin et al. (Derjaguin et al., 1947). In the 1980s and 90s, Anderson and co-workers (Anderson et al., 1982; Anderson and Prieve, 1984; Prieve et al., 1984; Ebel et al., 1988; Anderson, 1989) derived expressions for the mobility of diffusiophoretic particles in electrolytic and non-electrolytic solute gradients. Over the last two decades, the experimental research on diffusiophoresis has grown significantly. Broadly, this research can be divided into two main categories: passive diffusiophoresis and active diffusiophoresis. Passive diffusiophoresis involves particles moving in response to external concentration gradients of the solute. This is most commonly achieved in microfluidic devices where several studies have reported colloidal focusing (Abécassis et al., 2008; Shi et al., 2016; Shin et al., 2016; 2017c; Ault et al., 2017), i.e., the tendency



of colloids to concentrate in a particular region. In contrast, active diffusiophoresis refers to phenomena where the particle assists in creating solute concentration gradients, which has led to significant interest in developing autonomous micromachines (Palacci et al., 2013; Aubret et al., 2018).

Recent research on diffusiophoresis has demonstrated that it is crucial for a broad range of phenomena, such as exclusion zone formation near membranes (Florea et al., 2014), removal of contaminants during laundry (Shin et al., 2018), formation of patterns on vertebrate skins (Alessio and Gupta, 2023a), and possibly even migration patterns in humans Alessio and Gupta (2023b). In fact chemotaxis, i.e., movement of microorganisms upon sensing gradients in chemoattractants, is essentially a subclass for diffusiophoresis, and is mathematically identical to diffusiophoresis (Alessio and Gupta, 2023a; Alessio and Gupta, 2023b; Chu et al., 2021). From an application standpoint, diffusiophoresis has been used for creating living crystals (Palacci et al., 2013), micromotor machines (Aubret et al., 2018), membrane-less water filters (Shin et al., 2017c)

and low-cost zetasizers (Shin et al., 2017a). Therefore, given the breadth of the applications of this process from interfacial phenomena to colloidal physics and biophysical systems, this perspective seeks to share ideas on how diffusiophoresis can advance fundamental research in colloidal physics and enable new technologies in biomedicine and lab-on-a-chip.

In Section 2, we provide a brief summary of the fundamentals and applications of passive diffusiophoresis. In Section 3, we highlight the impact of particle shape and surface heterogeneity on active diffusiophoresis and discuss some of the promising applications. Finally, we suggest upcoming trends and outline future opportunities in Section 4.

2 Passive diffusiophoresis: A colloidal focusing mechanism

Definition: Passive diffusiophoresis is the movement of colloidal particles in response to external concentration gradients.

Figure 1 summarizes fundamental features of passive diffusiophoresis and recent developments in the field.

Physical explanation: Diffusiophoresis, in its simplest form, can be understood through excess osmotic pressure. Due to the interaction potential between the particle and the solute, the external solute concentration gradient and consequently the osmotic pressure gradient are enhanced near the particle surface. In the frame of reference of the particle, the fluid will have a tendency to move against the osmotic pressure gradients. However, since the fluid is stationary, the particle will move in the direction of osmotic pressure gradient, or towards higher solute concentrations; see Figure 1A. While the particle typically moves towards higher concentration of solute, it is possible that this direction can be reversed, for instance if there is electrostatic interaction between the solute and the particle (Velegol et al., 2016).

Mobility relationships: The derivation for diffusiophoretic mobility for a spherical particle has been covered extensively by Anderson et al. (1982), Prieve et al. (1984) and Keh and Wei (2000). Briefly, in these calculations, solute conservation equations and Stokes flow equations are solved simultaneously. Since the particles are small, the velocity of the particle is calculated by invoking a force-free condition such that the phoretic force is balanced by the Stokes drag. The calculations can be significantly simplified in the limit of shallow concentration gradient, small interaction potential and thin interaction limit. Several derivations exist for both electrolytic and non-electrolytic solutes. Recent work in the area focuses on diffusiophoresis in concentrated electrolytes (Prieve et al., 2019), multiple electrolytes (Chiang and Velegol, 2014; Gupta et al., 2019), dependence of mobility on solute concentration (Gupta et al., 2020; Lee et al., 2023b), multivalent electrolytes (Wilson et al., 2020), particle shape (Doan et al., 2023) and composite particles (Li and Keh, 2016; Mondal et al., 2023).

Observation in experiments: The most striking feature of passive diffusiophoresis is the “focusing” effect where a region of high colloid concentration is observed. This, in effect, is as if diffusiophoresis imparts a “negative diffusion” coefficient to colloids (Murray, 2003). This effect was observed by Staffeld and Quinn (1989), and has been repeatedly observed in various settings in recent literature (Shi et al., 2016; Shin et al., 2016; 2017c; Ault et al., 2017). Physically, this is understood from the fact that due to diffusiophoresis, particles have a tendency to migrate towards region of high solute concentration. However, their diffusion is significantly weaker due to their large size (compared to the solute) and thus they collect and move with the front of the diffusing solute and focus. We specifically highlight the work of Shi et al. (2016) who showed that the focusing can be controlled through an acid-base reaction; see Figure 1C left-panel.

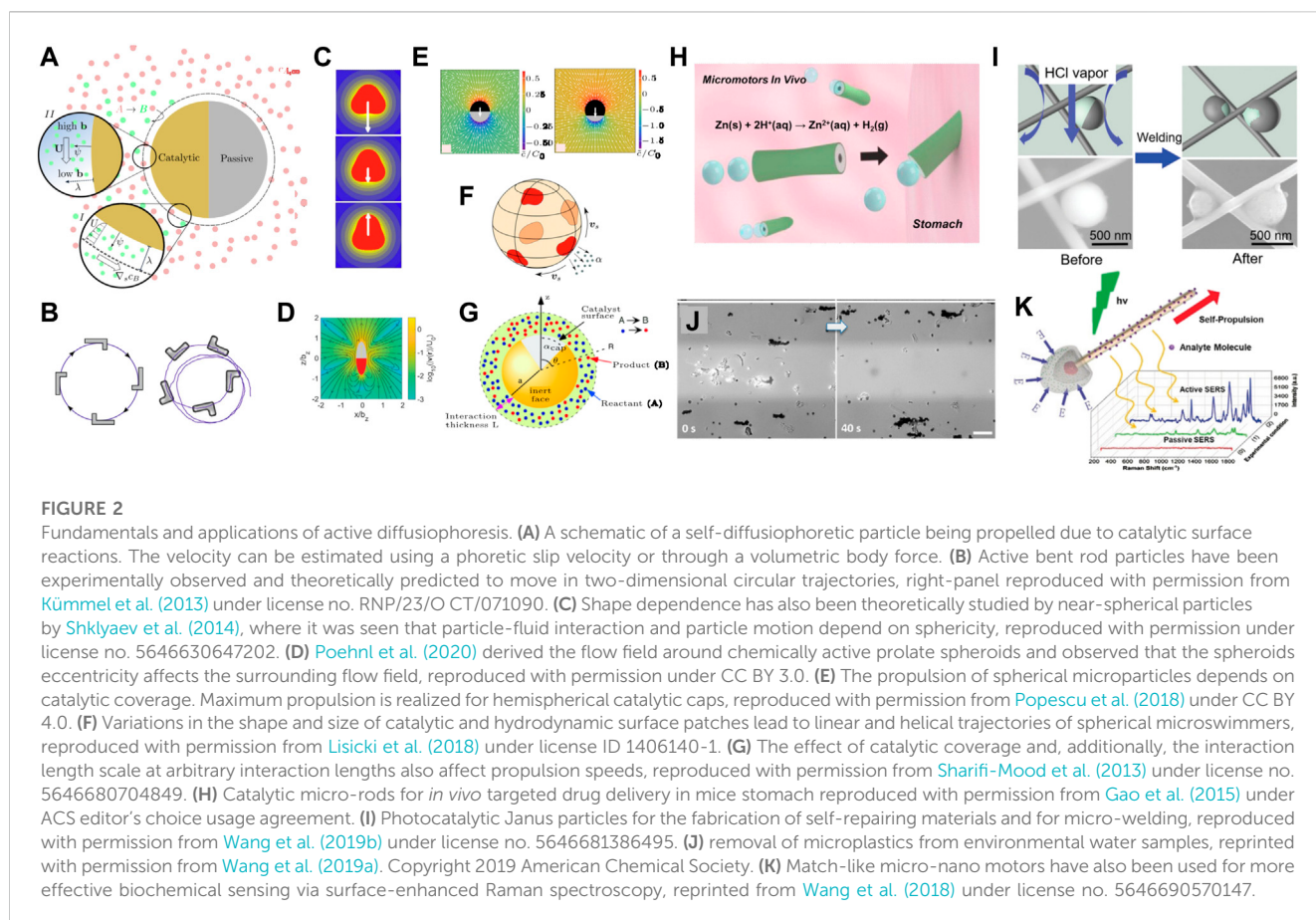
Diffusiophoresis and diffusioosmosis in tandem: Theoretical calculations based on diffusiophoresis alone for the setup described above significantly over-predict the focusing effect (Alessio et al., 2021), i.e., the experimentally observed focusing is weaker than theoretical predictions. Alessio et al. (2022) demonstrated that this is because of the diffusioosmotic flow generated by the sidewalls of the microfluidic channel and found that effective diffusivity of the particle is enhanced due to this flow, akin to Taylor dispersion (Figure 1B). They showed that this increased dispersion dampens the focusing and quantitatively recovered the experimental trends. The effect of diffusiophoresis and diffusioosmosis in tandem, combined with geometric and flow considerations, can be

employed to trap or eject particles (Kar et al., 2015; Shin et al., 2020; Singh et al., 2020; Singh et al., 2022a; Singh et al., 2022b).

Beyond one-dimensional gradients: The studies described above have largely focused on one-dimensional external gradients. This is due to the fact that appreciable two- and three-dimensional solute gradients are challenging to sustain because solutes diffuse quickly compared to the colloids they propel. Banerjee et al. (2016) proposed and demonstrated a creative mechanism for maintaining solute gradients in two dimensions by deliberately placing gel beacons which can act as solute sources or sinks, soaking them with solution, and flushing the arrangement with deionized water and colloids to observe long-lasting particle banding (Banerjee and Squires, 2019); see Figure 1C right-panel. Colloidal focusing via diffusiophoresis is still seen in these geometries, and the design principle is applied to more complicated situations such as particulate navigation in porous media (Tan et al., 2021). Recently, Raj et al. (2023b) extended this approach computationally by optimizing the spatial arrangement of arbitrary collections of sources and sinks for maximizing colloid concentration (Figure 1C right panel).

Diffusiophoresis-enhanced Turing patterns: An underexplored topic in diffusiophoresis literature is the use of reaction-diffusion instabilities. Alan Turing theorized in 1952 (Turing, 1952) that instabilities arising from non-linearities in chemical reaction pathways can be sufficient for sustained pattern formation and biological development from an initially homogeneous state. The fundamental mechanisms for biological pattern formation and the role of Turing instabilities to this day are still under scrutiny (Maini et al., 2012) and mechanisms such as motility-induced phase separation and chemotaxis are thought to be essential contributors (Cates and Tailleur, 2015; Zhao et al., 2023), though Turing patterns serve as an excellent starting point for analyzing spatial heterogeneity of chemical and biological materials (Figure 1D). Ramm et al. (2021) showed in experiments with pattern-forming *Escherichia coli* MinDE protein that unrelated DNA origami-streptavidin nanostructures, when dispersed over the already-patterned *E. coli* were organized via diffusiophoresis into mirroring patterns. This was the first-ever direct observation of diffusiophoresis in a cell biology context. The authors also discussed the focusing effect of diffusiophoresis wherein the patterns became sharper with larger cargo size (Figure 1E). Following this, Alessio and Gupta (2023a) developed a theory for this phenomenon and coined the term Diffusiophoresis-Enhanced Turing Patterns. The authors showed that diffusiophoretic focusing explains mysterious features such as sharpening and double-spotting of the chromatophores in vertebrae such as the Ornate Boxfish and the Jewel Moray Eel (Figure 1F) and can qualitatively recover the experimental results of Ramm et al. (2021). Inspired by the similarity between colloids and chemotaxis by the ubiquity of diffusiophoretic motion of biological microorganisms and colloids along gradients of chemical concentration, Alessio and Gupta (2023b) argued that humans also migrate towards concentration gradients of opportunities (economy, social connections, safety, etc.). Due to the focusing effect of diffusiophoresis, they were able to create hotspots in human population densities that mimic cities.

Applications: As is clear from the above discussion, the key feature of diffusiophoresis is the tendency to focus colloids. Since



high concentration provides an easy method to track colloidal fronts, diffusiophoresis has been suggested to create a low-cost Zetasizer (Shin et al., 2017a). Finally, given that diffusiophoresis can collect the particles to a particular region, it can also deplete the particles from a region, which enables it to clear contaminant from a fabric (Shin et al. (2018), see Figure 1G), and develop a membrane-less water filtration device (Shin et al. (2017c), see Figure 1H).

3 Active diffusiophoresis: The “chemical” mode of swimming

Definition: Active diffusiophoresis, also referred to as self-diffusiophoresis, is the movement of a particle in response to the concentration gradient of a solute it creates. Therefore, the particle plays an “active” role in its own propulsion. The most common approach to achieve active diffusiophoresis is to coat the particle surface with a catalytic patch (Golestanian et al., 2007), which subsequently creates concentration gradients due to a surface reaction. We refer to this mode of propulsion as “chemical” swimming (this could also include active droplets which are driven by surfactant concentration gradients which subsequently result in Marangoni forces (Dwivedi et al., 2022), but are not the focus of this perspective). This is distinct from the “mechanical” mode of swimming, where a particle propels by changing their shape with time, such as surface distortions (Stone and Samuel, 1996).

Physical explanation: The mechanism of active diffusiophoresis is similar to passive diffusiophoresis, i.e., osmotic pressure gradients help drive the motion. However, in self-diffusiophoresis, since the particle creates the gradient itself, asymmetry in particle geometry or catalytic patch is required to induce motion (Figure 2A). Therefore, the propulsion direction in active diffusiophoresis is dependent on the local orientation of particles, unlike passive diffusiophoresis. This feature can give rise to life-like collective behavior (Palacci et al., 2013).

Mobility relationships: The most common setup for self-diffusiophoresis is a spherical particle coated with a catalytic patch. The velocity (both translational and rotational) of the particle is typically calculated through one of two distinct approaches. In the first, the solute transport equation with a phoretic flux and a surface flux boundary condition is coupled to Stokes flow equations and solved by invoking the zero net force and torque conditions (Michelin and Lauga, 2014). In the second, a phoretic slip velocity is assumed on the particle surface and then the reciprocal theorem is invoked to obtain the translational and rotational velocity using the same zero net force and torque conditions (Stone and Samuel, 1996; Golestanian et al., 2007; Poehnl and Uspal, 2021; Ganguly and Gupta, 2023). The first approach is cumbersome but is more accurate because it can resolve the velocity as a function of interaction potential, interaction length scale, and Péclet number. On the other hand, the second approach is more popular but suffers from the limitation that it is only valid in the thin interaction limit and requires a

lumped mobility parameter to estimate the phoretic slip velocity (Figure 2A). In Ganguly et al. (2023), authors extended the second approach to develop a universal mobility relationship with a volumetric body force (Figure 2A) for spherical particles that reduces to relevant limits for both active and passive diffusiophoretic systems.

Shape and surface heterogeneity effects: One of the exciting features of self-diffusiophoresis is that the trajectories are orientation dependent and hence shape and surface heterogeneity serve as useful parameters to control trajectories. In Ganguly and Gupta (2023), authors focused on the geometry of a slender bent-rod. This geometry is particularly interesting because it can be described entirely by two dimensionless parameters: the length asymmetry of the two arms and the angle between the arms. The authors showed that the two-dimensional motion of a self-diffusiophoretic bent rod will always be circular, consistent with experimental reports (Kümmel et al. (2013); Figure 2B). Perhaps most importantly, the authors showed that even if the catalytic activity is uniform across the bent rod, the particle can self-propel entirely due to its geometric anisotropy. This was also observed in Shklyaev et al. (2014) where the authors described “chemical sailing” of nearly spherical particles. Specifically, the authors emphasize that even if the catalytic activity is uniform, the particles can swim entirely due to geometric anisotropy (Figure 2C). Poehnl et al. (2020) developed an analytical solution for a spheroidal swimmer (Figure 2D) and Poehnl and Uspal (2021) investigated a helical swimmer. For the latter, the authors developed a slender body theory, which is a useful method to investigate shape effects in active diffusiophoresis (Schnitzer and Yariv, 2015; Yariv, 2019; Katsamba et al., 2020). The realization of curvilinear trajectories in active-diffusiophoresis is not only limited to non-spherical particles. Such curvilinear motion could also be induced through surface heterogeneity (Sharifi-Mood et al., 2013; Lisicki et al., 2018; Raman et al., 2023). In fact, Lisicki et al. (2018) observed that both linear and helical trajectories can be obtained by controlling the placement of catalytic and hydrodynamic patches on a sphere (Figure 2F). Finally, the aforementioned features may be combined with interaction lengthscales to further change the mobility, as discussed in Sharifi-Mood et al. (2013).

Life-like or machine-like behavior due to collective motion: The applications of active diffusiophoresis may be broadly categorized into two different categories. In the first category, the focus is on collective behavior of particles. For instance, Palacci et al. (2013) showed how collective motion can create “living crystals” which assemble upon activation of surface reaction and disperse upon deactivation. In Aubret et al. (2018), it was shown that a similar setup can be used to create spinning microgears which can be activated and deactivated upon demand. The collective motion of active diffusiophoretic particles can thus be employed to create life-like or machine-like functionality.

Functionality due to autonomous motion of a single particle: In the second category of applications, the focus is instead on the autonomous motion of individual particles and how we can “steer” them on demand. There are several experimental studies exploring how these steerable microparticles could be used for biomedical applications such as targeted drug delivery (Gao et al., 2015; Peng

et al., 2015; Luo et al., 2020). Specifically, Gao et al. (2015) conducted *in vivo* experiments using catalytic micro-rods fueled by gastric juices in the stomach of mice; see Figure 2H. They found that catalytic actuation led to enhanced delivery and retention of drugs within the mucosal layer in the stomach. Beyond biomedical applications, self-diffusiophoresis is being employed in technologies related to environmental remediation, fabrication of functional materials, and chemical sensing. Das et al. (2015) studied how boundaries could be leveraged to steer active Janus particles, which Wang et al. (2019b) utilized to photocatalytically drive particles and weld junctions of Ag-nanowire networks; see Figure 2I. Similarly, Wang et al. (2019a) used photocatalytic active particles for removing microplastics from environmental water samples, see Figure 2J. Deng et al. (2022) reviewed recent work on behavior of active colloids in the vicinity of air-water interfaces, which is important to develop robust environmental remediation systems. Another application of self-diffusiophoretic particles is in chemical sensing, where catalytically driven particles chemically detect ricin and other toxins for bio-defense (Esteban-Fernández de Ávila et al., 2016), or enhance the effectiveness of existing biochemical sensing techniques such as SERS (Wang et al., 2018); see Figure 2K.

4 Future opportunities

Through this perspective, we sought to provide an overview of the crucial features of both passive and active diffusiophoresis. In particular, we emphasized that colloidal focusing underpins passive diffusiophoresis and enables a range of different applications. In contrast, in active diffusiophoresis, the particle shape and surface heterogeneity give rise to intriguing trajectories which are employed for autonomous swimming or collective behavior applications. Going forward, in our view, these are some of the future opportunities in the area of diffusiophoresis:

Electrochemical concentration gradients in passive diffusiophoresis: The concentration gradients typically employed in passive diffusiophoresis typically are externally imposed. However, employing electrochemical reactions to generate concentration gradients and subsequently using them as a driving force for diffusiophoresis remains relatively underexplored. Some recent studies have begun to investigate this phenomenon, referred to as electrodiffusiophoresis (Jarvey et al., 2022; Jarvey et al., 2023; Rath et al., 2021; Wang et al., 2022a; b). We believe this broad area provides opportunity for combining colloidal physics with electrochemistry and could be useful for directed assembly of colloids or removing deposited colloids on electrodes.

Porous media and charged confinement in passive diffusiophoresis: The majority of the work on passive diffusiophoresis focuses on unconfined geometries. Some preliminary experimental studies and theoretical investigations (Sambamoorthy and Chu, 2023) focus on confinement due to biofilms (Somasundar et al., 2023) or hydrogels (Doan et al., 2021). A few studies do consider diffusiophoresis in a charged cavity (Keh and Anderson, 1985; Zydney, 1995; Lee and Keh, 2014; Chiu and Keh, 2018), but future theoretical studies could focus on charged confinement in a capillary-like geometry, where double layers from the surface may interact with the double layers around the particles (Henrique et al., 2023; Henrique et al., 2022b;

Henrique et al., 2022a), and diffusioosmosis may also play a role. This is particularly important for diffusiophoresis of nanoparticles that may allow enhanced transport in nanoporous materials.

Shape and fluid flow effects in passive diffusiophoresis: Though shape has played a central role in active diffusiophoresis, it has been rarely investigated in passive diffusiophoresis. The recent work by Doan et al. (2023) et al. for ellipsoids shows the importance of particle orientation on trajectories. In addition, only a few studies have investigated background flow (Alessio et al., 2022; Alessio et al., 2021; Shin et al., 2016; Shin et al., 2017a; Shin et al., 2017b; Shin et al., 2020; Staffeld and Quinn, 1989) and recent studies have shown that it can significantly impact colloidal focusing (Chu et al., 2022; Chu et al., 2020). Therefore, future fundamental investigations can focus on particle shape and background flow. The knowledge from these studies will be particularly relevant for lab-on-a-chip applications.

Chemical swimming with other modes of swimming: Active diffusiophoresis provides a convenient way to control trajectories using shape and surface heterogeneity. However, these trajectories can be further controlled by combining two different modes of swimming. In Raj et al. (2023a), the authors investigated the combination of chemical and mechanical modes of swimming simultaneously, and noted that even reciprocal strokes yield nonreciprocal displacement when both modes are employed simultaneously. Similar investigations with other modes of activation such as electric fields and magnetic fields could provide improved control over particle trajectories (Lee et al., 2023a).

Combination of active and passive diffusiophoresis: Combining the two modes provides an exciting opportunity to exploit the two features described, i.e., diffusiophoretic focusing and orientation-dependent motion. This might enable a finer control on life-like crystal formation due to active control, for instance, to only occur in a certain region where particles focus due to passive control.

Applications: In addition to the applications described earlier, diffusiophoresis presents several opportunities for future applications. Similar to its role in laundry (Shin et al., 2018), diffusiophoresis can be used for cleaning membranes or removing particles from electrodes. Additionally, by triggering reaction-diffusion instabilities (Alessio and Gupta, 2023a), Turing patterns may allow one to sense external chemical concentration. Another possibility for sensing applications is to use passive and active diffusiophoresis simultaneously to create shape-shifting patterns in real-time for detection of harmful chemicals.

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Data availability statement

The original contributions presented in the study are included in the article/Supplementary material, further inquiries can be directed to the corresponding author.

Author contributions

AGa: Conceptualization, Writing–original draft, Writing–review and editing, Data curation. BA: Conceptualization, Data curation, Writing–original draft, Writing–review and editing. AGu: Conceptualization, Writing–original draft, Writing–review and editing, Supervision.

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Conflict of interest

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