Environmental Science and Ecotechnology 18 (2024) 100329

Contents lists available at ScienceDirect

Environmental Science and Ecotechnology

journal homepage: www.journals.elsevier.com/environmental-science-and-ecotechnology/

Piezoelectricity activates persulfate for water treatment: A perspective

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ARTICLE INFO

Article history:
Received 22 July 2023
Received in revised form
27 September 2023
Accepted 28 September 2023

Keywords:
Piezoelectric charge
Persulfate
Piezoelectric activation
Water treatment

ABSTRACT

Advanced oxidation processes (AOPs) utilizing persulfate (PS) offer great potential for wastewater treatment. Yet, the dependency on energy and chemical-intensive activation techniques, such as ultraviolet radiation and transition metal ions, constrains their widespread adoption. Recognizing this limitation, researchers are turning towards the piezoelectric effect—a novel, energy-efficient method for PS activation that capitalizes on the innate piezoelectric characteristics of materials. Intriguingly, this method taps into weak renewable mechanical forces omnipresent in nature, ranging from wind, tides, water flow, sound, and atmospheric forces. In this perspective, we delve into the burgeoning realm of piezoelectric/PS-AOPs, elucidating its fundamental principles, the refinement of piezoelectric materials, potential mechanical force sources, and pertinent application contexts. This emerging technology harbors significant potential as a pivotal element in wastewater pretreatment and may spearhead innovations in future water pollution control engineering.

1. Introduction

There is a growing recognition of the escalating influence of urbanization on the natural milieu, particularly in aquatic ecosystems [1]. In recent decades, the widespread utilization of chemical products across various domains, such as medicine, agriculture, livestock, and urban areas, has resulted in organic contaminants in water bodies [2], posing a threat to both surface and groundwater resources [3]. Therefore, it is an urgent need to develop cost-effective and highly efficient approaches for wastewater treatment. Advanced oxidation processes (AOPs) have emerged as a well-established technology for removing organic pollutants in water. These processes are characterized by generating highly reactive radicals that directly oxidize organic molecules into smaller molecules or even CO_2 and H_2O [4,5].

The appearance of persulfate (PS) in AOPs can be traced back to the early 21st century when Dionysiou's group first explored the cobalt-mediated decomposition of peroxymonosulfate (PMS) for organic pollutants degradation [6]. Distinct from conventional Fenton-based technologies, PS-AOPs, comprising PMS and peroxydisulfate (PDS), offer various technical and economic advantages for environmental applications. These include: (i) reduced safety risks and lower transportation costs, (ii) rich and diverse means of activating PS, (iii) less dependence on operating parameters such as pH and environmental medium, (iv) faster free radical generation and higher oxidation potential ($E^0(SO_4^{\bullet-}/SO_4^{2-})$ = +2.60 to +3.10 V_{NHE} > $E^0(HO^{\bullet}/OH^-)$ = +1.90 to +2.70 V_{NHE}) [7–9]. However, the existing activation methods toward PS, such as ultraviolet (UV), microwave, heat, and transition metal activation (Fig. 1a), necessitate either substantial energy consumption or continuous chemical input, significantly restricting their practical applications. Thus, the quest for environmentally friendly and energy-efficient activation techniques for PS remains an ongoing research endeavor [6,10].

The piezoelectricity effect manifests as an extraordinary electric response displayed by non-centrosymmetric crystalline materials upon exposure to mechanical force [11]. This unique interplay between mechanical and electrical energies bestows the capability for efficient conversion of environmental mechanical energies, such as wind, tide, water flow, sound, and atmospheric forces, into valuable electrical energy [12,13]. This unique behavior is rooted in the anisotropic nature of the crystal structure. When a piezoelectric material experiences external mechanical forces or strain, it prompts atomic displacements within the crystal lattice, leading to a mismatch between the positions of cation and anion centers. This disparity in atomic positions generates an inherent dipole moment, referred to as polarization. Consequently, the orderly summation of

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https://doi.org/10.1016/j.ese.2023.100329

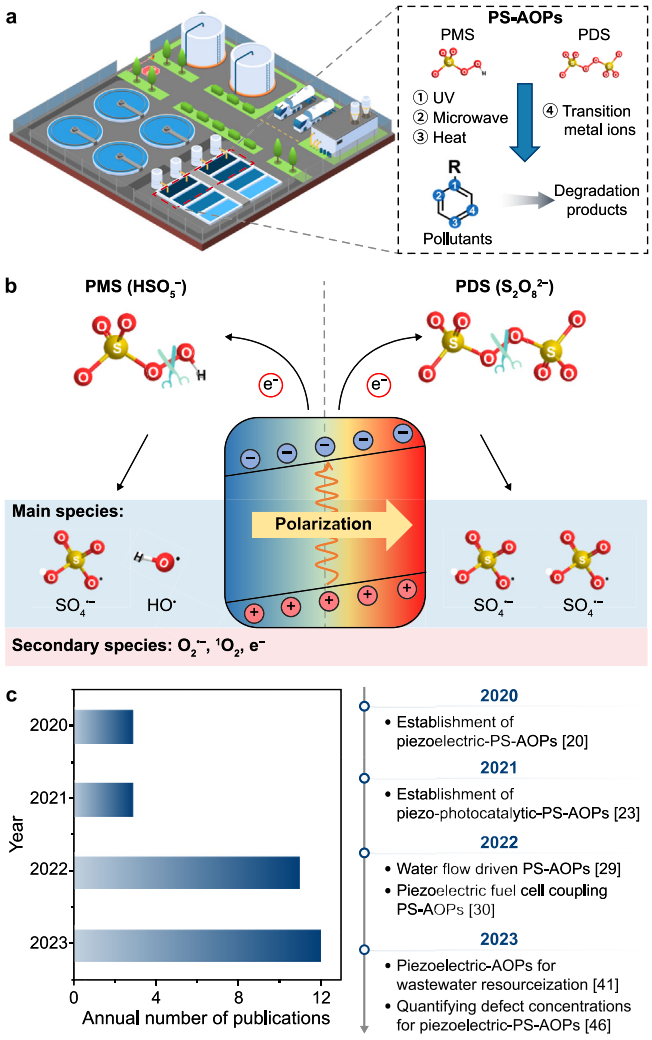

Fig. 1. **a**, Traditional activation method of PS-AOPs technology. **b**, Mechanism of piezoelectric/PS-AOPs. **c**, Annual number of publications concerning piezoelectric/PS-AOPs.

these dipole moments within the crystal cell engenders a macroscopic built-in potential field, commonly known as the piezoelectric potential [14]. A notable advantage of this piezoelectric potential lies in its resilience to applied strain or stress, as it arises from immobile and non-annihilating ionic charges [15]. These abundant localized charges serve as potent resources that can be harnessed for various catalytic reactions, including energy conversion and wastewater purification [16]. Intriguingly, these localized charges, brought forth by piezoelectricity, prove instrumental in disrupting the O—O bonds in PMS and PDS, facilitating activate PS for wastewater treatment [17,18].

Herein, our focus is directed towards piezoelectric/PS-AOPs, delving into its basic principles, piezoelectric materials and their improvement, sources of mechanical force, and potential application scenarios. Moreover, we systematically summarize and elucidate the great potential of this technology, underscoring its promising prospects as an integral component of AOP treatment systems. With these key objectives in mind, we strive to provide a comprehensive overview and offer insights into the future trajectory of piezoelectric/PS-AOPs. This perspective seeks to consolidate the current understanding of the field and chart a visionary course for forthcoming research directions in the years ahead, thereby boosting advancements in this emerging field.

2. Overview of piezoelectric/PS-AOPs

Our primary objective is to provide a comprehensive overview of the fundamental principles underpinning PS-AOPs. Specially, we delve into key areas such as piezoelectric materials and their improvement, the source of mechanical force, and application scenarios (Fig. 2). There remains an ongoing drive and selection process to identify and optimize key factors that drive the advancement of this technology.

Basic principles. The principal essence of PS-AOPs lies in the pivotal step of liberating $HO^\bullet$ and $SO_4^{\bullet-}$ through the cleavage of peroxide bond within the persulfate molecule. This peroxide bond scission in PMS or PDS can be achieved via electron transfer reactions during activation. As previously mentioned, the generation of localized charges through piezoelectricity plays a pivotal role in disrupting the O—O bonds in PMS and PDS, giving rise to a diverse array of active species, including main species ($SO_4^{\bullet-}$ and $HO^\bullet$) and secondary species ($O_2^{\bullet-}$, $^1O_2$, and e^-) (Fig. 1b). Specifically, when a piezoelectric material is subjected to an external mechanical force or strain, it induces displacement of the atoms within the lattice, resulting in a mismatch between the positions of the cation and anion centers. This displacement in atomic positions induces dipole moments that, when ordered and superimposed, can generate macroscopic built-in electric fields. These adequate polarized charges in the electric field can bring more free carriers to promote redox reactions, and this electric field can also drive the electron and hole pairs to migrate in the opposite direction under external force (equation (1)). The e^-_{piezo} tends to react with HSO_5^- ions to form $HO^\bullet$ (equation (2)) and $SO_4^{\bullet-}$ (equation (3)); or with $S_2O_8^{2-}$ to form $SO_4^{\bullet-}$ (equation (4)). Meanwhile, h^+_{piezo} can react with HSO_5^- to form $SO_5^{\bullet-}$ (equation (5)), and two $SO_5^{\bullet-}$ can form $^1O_2$ and $SO_4^{\bullet-}$ (equation (6)). On the other hand, a few of the piezoelectric-induced charges can react with the water molecules to form a small amount of $HO^\bullet$ (equation (7)), or with oxygen to form $O_2^{\bullet-}$

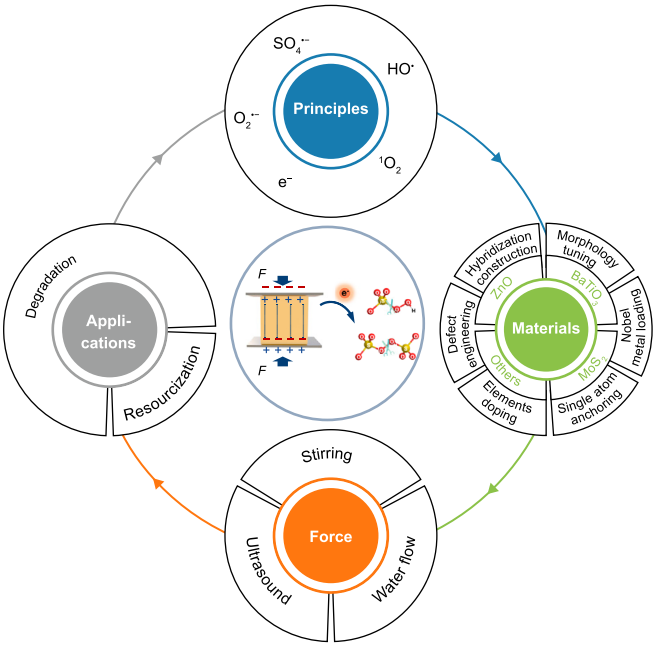

Fig. 2. Overview of piezoelectric/PS-AOPs: basic principles, piezoelectric materials and their improvement, source of mechanical force, and application scenarios.

(equation (8)). These resulting $O_2^{\bullet-}$ can react with $HO^{\bullet}$ to form $^1O_2$ (equation (9)). The specific activation process is shown as follows:

$$\text{Piezoelectric} + \text{Mechanical force} \rightarrow \text{Piezoelectric}\ (e^- + h^+) \quad (1)$$

$$HSO_5^- + e^- \rightarrow HO^{\bullet} + SO_4^{2-} \quad (2)$$

$$HSO_5^- + e^- \rightarrow OH^- + SO_4^{\bullet-} \quad (3)$$

$$S_2O_8^{2-} + e^- \rightarrow SO_4^{\bullet-} + SO_4^{2-} \quad (4)$$

$$HSO_5^- + h^+ \rightarrow H^+ + SO_5^{\bullet-} \quad (5)$$

$$SO_5^{\bullet-} + SO_5^{\bullet-} \rightarrow {}^1O_2 + 2SO_4^{\bullet-} \quad (6)$$

$$H_2O + h^+ \rightarrow H^+ + HO^{\bullet} \quad (7)$$

$$O_2 + e^- \rightarrow O_2^{\bullet-} \quad (8)$$

$$O_2^{\bullet-} + HO^{\bullet} \rightarrow OH^- + {}^1O_2 \quad (9)$$

The piezoelectric/PS-AOPs have garnered escalating attention in recent years, sparking a surge in research publications and an ever-growing interest in its application for environmental remediation (Fig. 1c). [17,19–46].

Piezoelectric materials and their improvement. Several materials available have been employed for the piezoelectric activation of PS. As the most extensively studied catalyst [17,19,20], $BaTiO_3$ was first successfully implemented in piezoelectric activation of PS for degradation of ibuprofen under ultrasound in 2020, which laid the foundation for numerous subsequent investigations on piezoelectric/PS-AOPs [20]. Other materials exhibiting asymmetric structures, such as ZnO and MoS_2, have also emerged as viable contenders for piezoelectric PS activation [32,45].

Nevertheless, various challenges persist with these piezoelectric catalysts, including inadequate capacity for harvesting mechanical energy, low surface area, and sluggish electron transfer rates. To address these obstacles, enormous efforts have been devoted to enhancing the performance of piezocatalytic PS activation, such as morphology tuning [20], noble metal loading [19], single atom anchoring [29], element doping [30,35,37,38], defect engineering [25,46], and hybridization construction [22,41]. For example, Xia et al. employed Ag-modified $BaTiO_3$ as a piezoelectric catalyst for activating PDS under ultrasonic vibration, leading to consecutive generation of $SO_4^{\bullet-}$ and $HO^{\bullet}$, effectively facilitating the removal of *Escherichia coli* (*E. coli*) [19]; Yang et al. successfully improved the utilization of PMS by quantifying the concentration of oxygen vacancies of ZnO, leading to highly efficient degradation of ornidazole (ORZ) [46]. The activation efficiency and decontamination performance of various piezoelectric materials were summarized in Table 1. These approaches seek to optimize the structural and compositional characteristics of the catalysts, thereby improving their piezoelectric response and catalytic activity in PS activation processes.

Source of mechanical force. The effectiveness of piezoelectric catalysis is profoundly influenced by the choice of mechanical force, which assumes a pivotal role in modulating the dipole field of the piezoelectric material. The piezoelectric potential, arising from the coherent summation of dipole moments within the material cells, can be amplified through external forces to drive the catalytic reaction.

In the laboratory, the mechanical forces employed for piezoelectric PS activation typically include stirring and ultrasound [34]. Stirring is often used for macroscopic mixing of liquids. However, it presents a challenge in achieving effective mixing and generating enough force to excite the material for piezoelectric effects. Ultrasound provides microscopic mixing of liquid materials and induces an ultrasonic cavitation effect by applying a periodic force that causes the rupture of cavitation bubbles. This leads to a high pressure of up to 10^8 Pa at the non-homogeneous catalyst/water interface, providing sufficient energy for electron excitation [16]. However, it is important to note that these activation methods necessitate electrical energy consumption to drive piezoelectric catalysis.

Looking ahead, envisioning the utilization of the subtle mechanical forces naturally present in the environment. In natural phenomena, weak mechanical forces hold a unique and intriguing status. These forces originate in many sources, including wind, tide, water flow, sound, and atmospheric forces. This weak mechanical force energy is tiny, yet its distinct advantage lies in its self-sustaining nature, relying solely on the energy provided by nature itself. Recent reports have presented exciting possibilities in activating PS through self-driven piezoelectric effects induced by water flow [29,32]. For instance, Lan and colleagues ingeniously harnessed water flow to drive Fe single atoms on the surface of MoS_2 for water purification, capitalizing on the latent mechanical forces in nature without any external energy input [29]. This groundbreaking discovery harnesses the inherent potential of weak mechanical forces and opens up a new avenue for piezoelectric catalysis applications. The exploration of other weak mechanical forces holds tremendous promise for future advancements in the realm of piezoelectric catalysis, expanding its scope and enhancing its compatibility with sustainable and eco-friendly practices.

Application scenarios. Nowadays, application scenarios of piezoelectric/PS-AOPs mainly include the degradation of pollutants and wastewater resource utilization. As previously discussed, the piezoelectric effect involves the generation of a polarized electric field within piezoelectric materials when subjected to an external mechanical force, inducing localized charges that activate PS for organic contaminants degradation [17]. Moreover, recent advancements have revealed that the piezoelectric potential can elicit long-range ordered driving forces, thereby facilitating the efficient separation of photogenerated electron-hole pairs across both the surface and bulk phases of materials [13,48]. Consequently, this pioneering discovery provides an extended approach to PS activation via piezo-photocatalysis for the degradation of pollutants.

Furthermore, the utilization of piezoelectric/PS-AOPs technology has recently showcased exciting prospects for the chemical conversion of organic pollutants into valuable chemical feedstocks with high conversion rates and selectivity of products other than CO_2. Simultaneously, it achieves efficient degradation of pollutants and production of H_2 during wastewater treatment [41,42]. Specifically, Xing's group prepared a Co_3S_4/MoS_2 catalyst under ultrasonic vibration to convert various pollutants to CO. The introduction of Co_3S_4 effectively promotes the transfer and utilization of piezoelectric electrons and shows a highly selective carbonate conversion to CO. The Co_3S_4/MoS_2/PMS system realized the selective generation of CO in practical and complex wastewater treatments, underscoring its potential for practical applicability [41]. They also used a MoS_2/Fe^0/PMS ternary system to achieve H_2 production and simultaneous degradation of organic pollutants in various wastewater. Notably, the ingenious incorporation of PS addresses the challenge posed by electron-consumptive inhibition of hydrogen production due to pollutants containing electron-absorbing groups [42]. These groundbreaking developments mark significant strides towards the realization of efficient and sustainable environmental remediation strategies.

Table 1
Different piezoelectric materials to activate persulfate for degrading contamination.

Sample	Activation force	Persulfate	Kinetic rate (min^{-1})	Contamination	Reference
$BaTiO_3$	Ultrasound	PMS	0.1002	Benzothiazole (BTH)	[17]
Ag−$BaTiO_3$	Ultrasound	PDS	-	*E. coli*	[19]
$BaTiO_3$	Ultrasound	PDS	0.0818	Ibuprofen (IBP)	[20]
MoS_2	Ultrasound	PMS	-	Phenol	[21]
$BaTiO_3$/MoS_2	Ultrasound	PMS	0.0559	ORZ	[22]
MoS_2	Ultrasound	PMS	0.0953	RhB	[25]
MoS_2	Ultrasound	PMS	0.129	Phenol	[26]
$SrBi_2B_2O_7$	Ultrasound	PDS	0.052	Sulfadiazine (SDZ)	[27]
CNTs/$BaTiO_3$	Stirring	PMS	0.021	Carbamazepine (CBZ)	[28]
Fe_1−MoS_2	Water flow	PMS	2.808	Metronidazole (MTZ)	[29]
$Bi_2Fe_4O_9$	Stirring	PDS	0.13	Bisphenol A (BPA)	[31]
MoS_2	Water flow	PMS	0.428	Benzotriazole (BTR)	[32]
$BaTiO_3$	Stirring	PMS	0.00815	CBZ	[34]
Co−$BaTiO_3$	Ultrasound	PMS	0.1245	Atrazine (ATZ)	[35]
Co−ZnAl-LDHs	Stirring	PMS	0.1644	Norfloxacin (NOR)	[37]
PVDF-MoS_2 membrane	Ultrasound	PMS	0.1704	CBZ	[39]
rGO/CN	Ultrasound	PDS	0.03509	Sulfamethoxazole (SMX)	[43]
α-$SnWO_4$/ZnO	Ultrasound	PMS	0.0177	CBZ	[44]
ZnO	Ultrasound	PMS	0.083	IBP	[45]
ZnO	Ultrasound	PMS	0.034	ORZ	[46]
Co−ZnO	Ultrasound	PDS	234	Levofloxacin (LVX)	[47]

3. The future of piezoelectric/PS-AOPs: from opportunity to challenge

Piezoelectric catalysis has emerged as a highly promising approach for PS activation, holding tremendous potential in environmental purification and wastewater resource utilization. In particular, it can decompose refractory organic pollutants in the aqueous environment, into CO_2 and H_2O or small molecules with low toxicity. Consequently, piezoelectric catalysis is poised to assume a pivotal role as a pre-treatment unit in AOP processes and a key component of future water pollution control engineering (Fig. 3). Its unique ability to harness the weak mechanical forces present in nature, including wind, tides, water flow, sound, and atmospheric forces, sets it apart from traditional energy- and resource-intensive methodologies, positioning it as a transformative force in the field of AOP technology.

Piezoelectric activation of PS is still in its early stages, with many underlying mechanisms and activation modes remaining underdeveloped. To propel the advancement of piezoelectric/PS-AOPs, dedicated efforts in the following areas are crucial:

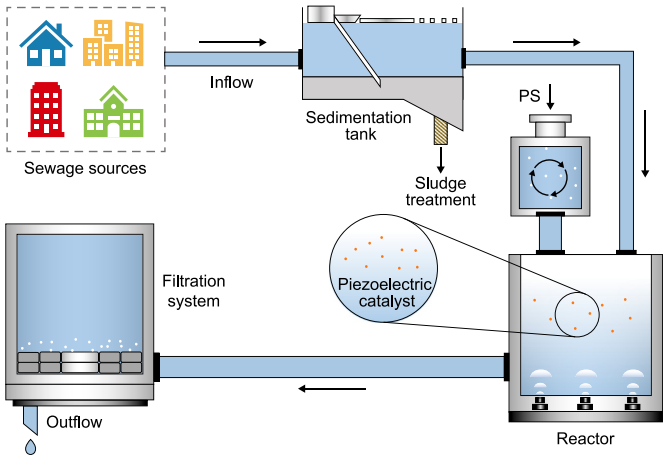

Fig. 3. Expanded future applications of piezoelectric/PS-AOPs for water treatment.

(i) Develop more efficient piezoelectric materials for PS activation. Exploring additional materials exhibiting inherent asymmetry for piezoelectric activation of PS holds significant promise. Widening the scope beyond $BaTiO_3$, MoS_2, and ZnO and delving into other two-dimensional (2D) materials with inherent structural asymmetry may unveil novel catalysts with improved piezoelectric response and catalytic activity [49].

(ii) Optimize the external forces of piezoelectric/PS-AOPs. As previously elucidated, current piezocatalytic reactions primarily rely on ultrasound as an external force. However, this activation mechanism arises from coupling multiple effects, including ultrasonic cavitation, piezocatalysis, flexoelectric effect, contact-electro-catalysis effect, and energy band theory. Consequently, clarifying these effects becomes challenging, hindering a comprehensive understanding of the underlying mechanisms governing piezoelectric catalysis. Stirring, water flow, and other methods also face the same challenges. Therefore, exploring innovative approaches to apply piezoelectric external forces effectively is essential, as it promises to unravel the intricate reaction mechanisms of piezoelectric catalysis.

(iii) Modulation of active species production in piezoelectric/PS-AOPs. The active species of PS-AOPs are diverse, including free radicals($SO_4^{\bullet-}$, $HO^{\bullet}$, and $O_2^{\bullet-}$) and non-free radicals ($^1O_2$ and electron transfer). The radical species produced are both electrophilic and nucleophilic, manifesting relatively stable reactivity without distinct selectivity. Conversely, the non-radical species generated exhibit electrophilic properties, affording controlled reactivity strength and certain reaction selectivity. Recent studies have highlighted the potential of innovative material design strategies and other approaches to exert precise control over the generation of active species [50]. These show promise in the future for directing the reaction pathway to obtain the desired product, thereby improving the selectivity and efficiency of PS piezocatalytic activation to cope with the complexity of organic pollutants in real water bodies.

(iv) Expansion of practical applications. The degradation of organic pollutants in real water remains an unresolved issue, owing to the high energy consumption of activation methods and multi-pollutants. The successful use of water flow self-

driving forces in piezoelectric/PS-AOPs offers great potential for wastewater treatment, suggesting the promise of piezoelectric/PS-AOPs in urban pipe drainage in the future.

CRediT authorship contribution statement

Zhi Li: Conceptualization, Investigation, Writing - Original Draft. **Shenyu Lan:** Writing - Review & Editing, Funding Acquisition. **Mingshan Zhu:** Conceptualization, Supervision, Writing - Review & Editing, Funding Acquisition.

Declaration of competing interest

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

Acknowledgements

This work has been supported by the National Science Foundation of China (No. 22322604 and 22006052), the Guangdong Basic and Applied Basic Research Foundation (No. 2020B1515020038), and the Pearl River Talent Recruitment Program of Guangdong Province (2019QN01L148). Some material in Fig. 1a is designed by macrovector/Freepik.

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