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Concentrating Immiscible Molecules at Solid@MOF Interfacial Nanocavities to Drive an Inert Gas-Liquid Reaction at Ambient Conditions

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Abstract: Gas-liquid reactions form the basis of our everyday lives, yet they still suffer poor reaction efficiency and are difficult to monitor in situ, especially at ambient conditions. Herein, we drive an inert gasliquid reaction between aniline and CO₂ at 1 atm and 298 K by selectively concentrating these immiscible reactants at the interface between metal-organic framework and solid nanoparticles (solid@MOF). Real-time reaction SERS monitoring and simulation investigations affirm the formation of phenylcarbamic acid, which was previously undetectable because they are unstable for post-reaction treatments. The solid@MOF ensemble gives rise to a >28-fold improvement to reaction efficiency as compared to ZIF-only and solidonly platforms, emphasizing that the interfacial nanocavities in solid@MOF are the key to enhance gas-liquid reaction. Our strategy can be integrated with other functional materials, hence opens up new opportunities for ambient-operated gas-liquid applications.

Gas-liquid reactions are crucial to sustain human civilization. They participate in various biological functions, such as photosynthesis¹ and gas exchange in bloodstream,² and also industrial processes,³ including hydrogenation of unsaturated compounds and gas scrubbing.⁴ Notably, the key principle to drive high-performance gas-liquid reactions is to enforce efficient and frequent intermolecular collisions between these immiscible phases. However, such reactions still suffer poor efficiency and tracking gas-liquid reaction in situ is challenging because of low gas concentration, especially at ambient/low pressure conditions. These performances are further aggravated by poor gas solubility and comparatively lower density of gas,⁵ whereby gas rapidly escapes a liquid medium and impedes efficient intermolecular interaction. To address these issues, conventional strategies involving flow chemistry,⁶ packed column and microbubbles have been developed to promote gas-liquid interactions by prolonging gas flow in liquid,⁷ or by increasing gas concentration/energy at >2 bar and >323 K.8 While these strategies have improved gasliquid reactions, they are limited by the need of energy-intensive and/or long reaction procedures to drive the reaction, even at high temperature/pressure. Operating gas-liquid reaction at ambient conditions remains a formidable challenge.

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Metal-organic framework (MOF) is an emerging porous material that could address these limitations through their excellent sorptivity and selectivity toward small gas/liquid molecules.9 When coated over a solid surface, the solid@MOF ensemble contains nanoscopic cavities at the solid-MOF interface.¹⁰ Gas sorption by solid@MOF platform continuously accumulates gas molecules in these confined cavities, eventually creating a pseudo high-pressure microenvironment even under ambient operations.¹⁰ We hypothesize that solid@MOF platform could also promote efficient gas-liquid reaction by concentrating biphasic molecules into close proximity to enhance their intermolecular interactions. Besides enhancing a reaction, molecular enrichment in the interfacial cavities of a plasmonic solid@MOF platform will also enable ultrasensitive in situ reaction monitoring via surface-enhanced Raman scattering (SERS). This molecular-level information is essential to elucidate reaction dynamics and facilitate rationale design of gas-liquid reaction toward optimal performance, efficiency and ambient operation.

Herein, we drive an inert gas-liquid reaction at ambient conditions by integrating a gas-sorbing layer of zeolitic imidazolate framework-8 (ZIF) over an array of Ag nanocubes (Ag@ZIF). Our integrated platform involving ZIF (pore aperture 3.4 Å) selectively concentrates and forces the immiscible gaseous and liquid-based reactants together to promote molecular interactions at the solid-MOF interface (Figure 1A). Moreover, Ag also functions as a SERS platform to track gas-liquid reaction in situ at the molecular level. Our model gas-liquid reaction is the reaction between liquid aniline and gaseous CO2 to form phenylcarbamic acid,9 an industrially significant process that traditionally requires high operating pressure/temperature even in the presence of catalyst. We first investigate the selective enrichment of liquid aniline within Ag@ZIF's interfacial cavity, which is vital for subsequent reaction with CO2 gas. Corroborating experimental SERS and density functional theory (DFT) studies, we track gas-liquid reaction in situ and demonstrate the unprecedented reaction of aniline and CO₂ at 1 atm and 298 K. Moreover, we reveal that the presence of interfacial cavities is the key to improve gas-liquid reaction efficiency by >28-fold which cannot be achieved in standalone ZIF or Ag platforms. Our work offers valuable insights to realize ambient-operated gas-liquid reactions, which are highly sought after in diverse fields including air-to-fuel conversion, chemistry, environmental remediation and industrial chemical manufacturing.

We begin by depositing an array of Ag nanocubes (edge length, 126±5 nm; Figure S1) on a Si substrate via Langmuir-Blodgett technique. The Ag nanocube array is then surfacegrafted with perfluorodecanethiol (PFDT) to provide a featureless SERS background in the region of 400 to 1800 cm⁻¹ necessary for accurate reaction monitoring. PFDT-grafted Ag nanocube array is then coated with a ZIF layer via repeated immersions into



Figure 1. (A) Schematic depicting the importance of Ag@ZIF to capture and concentrate gas and liquid molecules in its interfacial cavities for enhanced gas-liquid reaction, using the reaction between CO_2 and aniline as a model. (B) Cross-sectional SEM image and (C) XRD pattern of Ag@ZIF. (D) Schemes and SERS spectra of (i) Ag@ZIF and (ii) Ag-only platforms when submerged in an aniline/decane mixture. (E) Time-dependent SERS intensity of aniline measured using Ag@ZIF or Ag-only platforms.

methanolic zinc nitrate and 2-methylimidazole mixture. ZIF nucleates and eventually grows into a polycrystalline film that fully encapsulates the Ag nanocubes (Ag@ZIF; ZIF thickness, 241±45 nm; Figure 1B; Figure S2). X-ray diffraction (XRD) analyses highlight the successful formation of Ag@ZIF platform (Figure 1C), where Ag@ZIF contains the unique XRD patterns of both ZIF and Ag nanocube. All Ag@ZIF platforms are activated using heat treatment prior to subsequent experiments (Figure S2-3).

There are two key criteria for solid@MOF to facilitate gasliquid reaction - its abilities to (1) selectively sorb gas and liquid species from reaction medium and (2) subsequently enrich these reactants at solid-MOF interface. To demonstrate its molecular selectivity and concentrating effects, we monitor the molecular components in solid@MOF interfacial cavity by immersing 0.1 M aniline in decane solution under N_2 environment using SERS. Only vibrational fingerprint of aniline is observed throughout the experiment, with SERS bands at 1274, 1387, 1594 and 1615 cm⁻ ¹ indexed to (v_{CN}), (ρ_{NH2} +v_{CC}), (δ_{NH2} +v_{CC}) and (v_{CN}+ δ_{NH2} +v_{CC}), respectively (Figure 1D, E).11 The absence of other bands indicates that decane is not accessible to SERS surface, and also eliminates the possibility of laser-induced processes on Ag@ZIF platform and/or aniline (Figure S4). Aniline's SERS intensity at 1387 cm⁻¹ gradually increases from (31±3) counts at 0 min to (463±16) counts within the first 90 min, followed by a plateau at ~466 counts beyond 105 min. The initial rise and subsequent



Figure 2. (A) SERS spectra and time-resolved contour plot recorded from Ag@ZIF (i) before reaction, (ii) after reaction and (iii) upon 30 min N₂ flush after reaction. (B) Temporal changes to SERS intensities of 1387 cm⁻¹ and 1427 cm⁻¹ bands when Ag@ZIF is submerged in 0.1 M aniline/decane mixture.

plateau of aniline's SERS intensity indicates the continuous accumulation and saturation of aniline near the SERS surfaces, respectively. Ag@ZIF exhibits a Lagergren pseudo first-order sorption kinetics model with a rate constant of 37.5 ms⁻¹, denoting the rapid sorption of aniline (Figure S4B). Conversely, no aniline SERS band is observed in control Ag platform without ZIF. Instead, we only observe three weak SERS bands at 1301, 1377 and 1448 cm⁻¹ attributed to decane's T_{CH2} , ω_{CH2} and δ_{CH2} ,¹² respectively (Figure 1D, E; Figure S5). These comparisons demonstrate the importance of creating a crack-free ZIF membrane to selectively capture and enrich target molecules over the encapsulated SERS-active surfaces. The selective sorption of aniline over decane arises from ZIF's characteristic pore size (3.4 Å) and framework flexibility to enable the diffusion of aniline (~5.9 Å) into SERS-active region,¹³ while blocking the access of larger solvent molecules such as decane (>7.2 Å). Furthermore, the rationale choice of solvent is another critical factor to ensure the selective capturing of reactant from liquid medium; the use of smaller solvent molecules (e.g. methanol) leads to competitive sorption with aniline by Ag@ZIF (Figure S6).

We subsequently drive an inert gas-liquid reaction between CO₂ and aniline at ambient conditions and concurrently monitor the reaction *in situ* using our Ag@MOF platform. In a typical reaction set-up, we first immerse Ag@ZIF in aniline/decane mixture for 2 hours to preconcentrate aniline in the interfacial cavities. Gas-liquid reaction is then initiated by bubbling CO₂ gas (5 sccm, 1 atm) into the reaction cell, and we track its progress in real-time using SERS. At 0 min, the SERS spectrum shows only aniline's vibrational signatures at 1274 (v_{CN}), 1387 (ρ_{NH2} +v_{CC}), 1594 (δ_{NH2} +v_{CC}) and 1615 cm⁻¹ (δ_{NH2} +v_{CC}; Figure 2A). As CO₂ exposure increases, aniline SERS intensity decreases

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exponentially at 15 min and eventually diminishes by 45 min (Figure 2A, B; Figure S7). We also note the emergence of new SERS bands at 1335, 1427, 1556 and 1633 cm⁻¹ at 30 min, whereby the intensities increase and ultimately dominate the SERS spectrum at 120 min. More importantly, subsequent purging with inert N₂ gas for 30 min does not revert these spectral evolutions. Our findings evidently demonstrate the spectral changes are due to irreversible chemical transformation of aniline. In contrast, control Ag platform in the absence of ZIF coating or Ag@ZIF under inert N₂ gas flow display no distinct spectrum changes (Figure S4, S7). Both control experiments highlight that ZIF is critical to enable gas-liquid reaction at ambient conditions and CO₂ functions as a gas reactant, respectively. The ZIF layer remains stable throughout the reaction with consistent thickness and XRD patterns before and after reaction (Figure S2-3).

We use density functional theory (DFT) to identify the chemical species involved in the reaction and elucidate the molecular origins behind the observed spectral evolutions. We simulate the SERS spectra of various probable carboxylated products including phenylcarbamic acid, phenyl phenylcarbamate, 1,3diphenylurea and 2-aminobenzoic acid owing to the possibility of carbamates/urea formation as well as the presence of orthodirecting -NH₂ group (Figure 3A, B; Figure S8). Experimental and DFT-simulated SERS spectra of aniline (Figure S9; Table S1)¹¹ are in close agreement, thus validating the accuracy of our DFT investigations. Most importantly, the experimental SERS spectrum at 90 min matches well to the simulated spectrum of phenylcarbamic acid. This is evident from the presence of unique amide and hydroxyl vibrational modes, whereby SERS features at 1427, 1335 and 1556 cm⁻¹ are indexed to $(v_{as,NC(OH)}+\delta_{COH}+v_{CO})$, $(v_{NCO}+v_{as,CCN})$ and $(v_{NCO}+v_{as,CNC}+\delta_{CNH})$; Figure 3B; Table S2), respectively. The formation of phenylcarbamic acid is also supported by similar work reporting the formation of carbamic acid using amine species and CO₂.¹⁴ We exclude other carboxylated species as the product owing to large spectral mismatch between our experimental and DFT-simulated SERS spectra (Figure S8).

Collectively, our experimental and DFT SERS studies clearly provide the first experimental verification to the formation of phenylcarbamic acid using liquid aniline and CO₂ gas (Figure 3C). In contrast, phenylcarbamic acid cannot be identified using conventional ex situ monitoring methods because it is unstable toward post-reaction treatments, such as molecular extraction. By enabling in situ gas-liquid reaction read-out at ambient conditions, our SERS-active Ag@ZIF platform addresses the limitations of modern characterization methods, which are typically nonmolecular-specific and/or mandate pressurized system. Moreover, the realization of such gas-liquid reaction is unprecedented at ambient conditions; most carbamate-forming reactions were typically performed at high temperature and pressure. Our ability to drive inert gas-liquid reaction and perform real-time reaction monitoring are enabled by Ag@ZIF's abilities to selectively concentrate target liquid and gas reactants within its interfacial cavities and near the plasmonic surfaces. This molecular concentrating process consequently forces biphasic reactants into proximity, thereby enhancing their intermolecular interactions to drive chemical reactions in the absence of sophisticated set-up and/or elevated pressure/temperature.



Figure 3. (A) Molecular structures of aniline and phenylcarbamic acid near Ag surface. (B) Key spectral transformations involving amide and hydroxyl moieties. (i) Experimental SERS spectra at 0 and 90 min. (ii) Simulated SERS spectra of aniline and phenylcarbamic acid. (C) Scheme depicting the gas-liquid reaction between aniline and CO_2 to form phenylcarbamic acid in the interfacial cavity.

Utilizing the critical reaction insights gained from our SERS and DFT investigations, we further apply Ag@ZIF platform at a larger reaction scale to demonstrate its versatility and scalability. Note that Ag@ZIF core-shell nanoparticles (2 mg; ZIF thickness. 47±11 nm; Figure S10-11) are used in this study, instead of Ag@ZIF substrate. This is to maximize their interactions with gas and liquid reactants throughout the reaction volume. We suspend Ag@ZIF particles in 0.1 M aniline/decane mixture and initiate the reaction by bubbling CO₂ gas. The reaction is monitored over 5 hours using UV-Vis absorption spectroscopy. We observe aniline's characteristic absorption peak at 234 nm (Figure 4A). With increasing CO₂ exposure from 0 min to 300 min, C/C₀ decreases linearly from 1 to 0.765 and corresponds to a reaction efficiency of 23.5% (Figure 4B). In contrast, control Ag-only platform and blank (neat aniline/decane mixture) exhibit an apparent efficiencies of <6% (Figure S14), which are likely due to minor adsorption of aniline on reaction apparatus. The >4-fold improvement to reaction efficiency in Ag@ZIF eliminates the possibility of catalytic effect arising from Ag or potential side reactions as the main contributor to its superior reaction performance. More importantly, Ag@ZIF exhibits >2-fold superior reaction efficiency compared to pure ZIF-only crystallites (2 mg) with a reaction efficiency of 11.7% (Figure 4B; Figure S13-14). By further normalizing the aniline consumed with the mass of ZIF (Figure 4C; Supporting Text 1), we ensure a fair comparison between Ag@ZIF and ZIF-only platforms to highlight the former's ability to facilitate gas-liquid reaction at the interfacial nanocavities. The Ag@ZIF exhibits an aniline consumption of ~9.6 $mg_{aniline}$ mg_{ZIF}⁻¹ over 5 hours, which is notably >28-fold higher than ZIF's consumption of ~0.3 mg aniline mg ZIF⁻¹. Ag@ZIF's superior reaction performance is also supported by its 2-fold higher apparent reaction rate constant (k) of 4.8 M ms⁻¹ as compared to ZIF-only

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Figure 4. (A) Aniline extinction spectra at predefined timings in presence of Ag@ZIF. Inset is a TEM image of our core-shell Ag@ZIF platform. (B) Changes to aniline's C/C_0 under prolonged CO_2 exposure. Blank refers to absence of any nanoparticles. (C) Mass of aniline consumed per unit mass of ZIF in Ag@ZIF and ZIF-only platforms. (D) Mass of aniline consumed per unit mass of ZIF and interfacial area in Ag@ZIF and SiO₂@ZIF systems.

platform (k = 2.4 M ms⁻¹; Figure 4B). These observations highlight the enhanced gas-liquid reaction in Ag@ZIF is primarily due to the presence of interfacial cavities rather than intrinsic ZIF pores.

Our strategy building on solid@MOF's interfacial cavities to promote gas-liquid reaction is also easily extended to other functional material. As a proof of concept, we synthesize SiO₂@ZIF (ZIF thickness, 14±4 nm; Figure S12) and subject them to identical reaction procedures. Using aniline's absorption peak at 234 nm, SiO₂@ZIF demonstrates a reaction efficiency of 20% (Figure S13) at 300 min, which is also ~3-fold higher compared to control SiO₂-only and blank systems (<7% efficiency). This again implies negligible contribution from the encapsulated solid to the decrease in aniline concentration. We further normalize the aniline consumption in both Ag@ZIF and SiO2@ZIF against interfacial area to exemplify that our strategy is independent on the nature of encapsulated solid. Both SiO2@ZIF and Ag@ZIF platforms exhibit similar aniline consumption of ~90.9 and 99.7 $mg_{\text{aniline}}.mg_{\text{ZIF}}{}^{-1}.\mu m_{\text{interfacial}} \quad {}_{\text{area}}{}^{-2}, \quad respectively$ (Figure 4D: Supporting Text 2). This is a direct evidence that the interfacial cavities between MOF and any solid materials is the key factor to achieve high-performance gas-liquid reaction. Our solid@MOF strategy is versatile and can be potentially integrated with many functional materials, such as solid catalyst, to promote diverse fluid-fluid reactions.

In conclusion, we drive an inert gas-liquid reaction between aniline and CO_2 at ambient conditions and realize its *in situ* reaction monitoring by selectively concentrating these biphasic molecules in the interfacial nanocavities of solid@MOF platform. Experimental and DFT SERS investigations jointly highlight the formation of phenylcarbamic acid in our model reaction. Notably, such molecular-level identification of phenylcarbamic acid is unprecedented because this chemical species is unstable to postreaction treatment and can only be tracked in situ. Moreover, solid@MOF improves reaction efficiency by >28-fold as compared to solid-only or MOF-only platforms, highlighting that the interfacial cavities present in solid@MOF is the origin behind its superior reaction performance. Such solid@MOF system tackles the long-standing bottleneck in gas-liquid reaction/monitoring by enriching immiscible reactants locally without requiring elevated temperature/pressure operations. Together with its high versatility to integrate with other functional materials, we envisage our solid@MOF strategy as a cornerstone realize efficient and ambient-operated fluid-fluid to reactions/processes critical in diverse fields including chemistry, heterogeneous catalysis, greenhouse gases removal and gas-tofuel conversions.

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