

Sub-nanoparticles crosslinking network organogel as the underwater adhesive

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ABSTRACT: Adhesives are applied extensively in daily life, and due to performance failure of conventional polymer adhesives in underwater environments, there is an urgent demand for highly stable underwater adhesives to meet application requirements of stable adhesion in humid environments and rapid leak sealing. Herein, crosslinking network of aluminum oxo clusters sub-nanoparticles (CN ASNPs) is prepared on a large-scale through a facile stirring reaction at room-temperature and atmospheric pressure, showing good gelation properties in non-polar solvents. The organogel exhibits outstanding underwater adhesive performance, arising from synergy effect of hydration layer disruption, micro-nano mechanical interlocking, high polymerization degree and intermolecular forces. And through being modified with perfluorodecyltriethoxysilane (FDETS) and methyltrimethoxysilane (MTMS), the adhesive performance can be further improved based on enhanced hydrophobicity and cohesion. The CN ASNPs organogel adhesive is suitable for various substrates, and for steel, the underwater adhesive strength can achieve ~ 2.41 MPa. More importantly, this adhesive exhibits excellent stability, whether in tap water or saltwater, whether under changed temperature (25–90 °C) or prolonged time (~ 180 days) conditions. And this adhesive will not cause any damage to substrates or leave hard-to-remove residues after bonding. Overall, the CN ASNPs organogel adhesive has great potential for practical applications in underwater and humid environments.

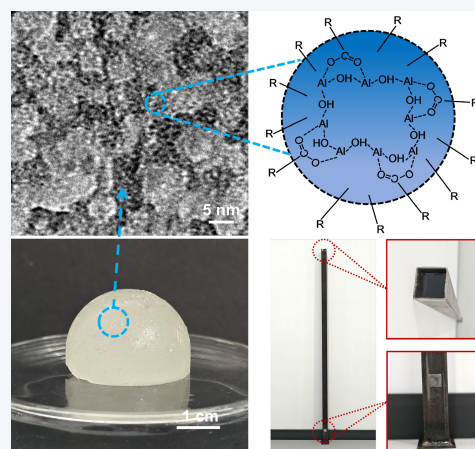
KEYWORDS: sub-nanoparticles, organogel, underwater adhesive, hydrophobic modification

1 Introduction

Adhesives play an indispensable role in modern industrial production and daily life due to their ability to efficiently bond materials through interfacial adhesion and cohesive strength [1–4]. They are widely applied in various fields such as electronic devices [5–7], automotive manufacturing [8], medical equipment [9–11], and construction engineering, providing reliable structural support and functional integration for various products. With the expansion of application scenarios to complex humid environments including

marine engineering [12, 13], underwater repair [14–19], biomedicine [20–25], and underwater sensors [14, 26–30], the demand for high-performance, high-stability underwater adhesives have become increasingly urgent. Underwater adhesives must overcome unique challenges posed by the aquatic environment, making them a research hotspot and technical difficulty in the field of adhesive materials [31–34].

The achievement of stable underwater adhesion performance is severely restricted by the intrinsic characteristics of the aquatic environment: On the one hand, water molecules are prone to adsorb on the substrate surface to form a dense hydration layer, which hinders direct contact between the adhesive and the substrate, thereby weakening interfacial interactions [33, 35, 36]; on the other hand, long-term immersion in water can cause plasticization, swelling, and even hydrolysis of the adhesive itself, leading to a decrease in cohesive strength and ultimately the failure of the bonding interface [37–41]. In addition, water molecules can



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disrupt interfacial forces such as hydrogen bonds and electrostatic forces, further reducing adhesive strength and long-term stability. Despite significant progress in the development of underwater adhesives over the past few decades, including marine organism-inspired biomimetic strategies (catechol chemistry) [32, 39, 42, 43] and material designs based on hydrogen bonding [44, 45], metal coordination [46–48], host–guest interactions [12, 49, 50], and electrostatic interactions [51, 52], existing materials still have prominent limitations. For some emerging mussel-inspired protein-based adhesives and supramolecular adhesives, precise and sophisticated structural design is required [53], which increases the preparation costs. For most underwater adhesives currently available, their limitations also include: insufficient adhesive strength in underwater environments [12, 20], poor tolerance to extreme temperatures (ultra-low or high temperatures) [12, 21], complex synthesis processes [18, 43], high raw material costs [18, 19, 26], and difficulty in large-scale production. Furthermore, some adhesives may cause damage to substrates or leave hard-to-remove residues after bonding, limiting their practical applications.

In this work, crosslinking network of aluminum oxo clusters subnanoparticles (CN ASNPs) is synthesized via a facile stirring reaction at room-temperature and atmospheric pressure, which can be produced on a large-scale, laying the foundation for practical applications. Because of the ultrahigh specific surface area, high polymerization degree and significant intermolecular interactions, the as-prepared CN ASNPs can form a robust three-dimensional (3D) network structure when dispersed in non-polar solvents, such as cyclohexane, and based on this, the CN ASNPs show good gelation properties. The CN ASNPs organogel exhibits outstanding underwater adhesive performance, arising from synergy effect of hydration layer disruption, micro-nano mechanical interlocking, high polymerization degree and intermolecular forces. Furthermore, the CN ASNPs organogel adhesive is modified with perfluorodecyltriethoxysilane (FDETS) and methyltrimethoxysilane (MTMS). Owing to the enhanced hydrophobicity promoting disruption of the hydration layer, the increased hydrogen bonds strengthening the interaction between the adhesive and substrates, and the hydrolysis induced polycondensation enhancing cohesion, the adhesive performance is successfully improved. The CN ASNPs organogel adhesive is applicable to various substrates. For the steel, it exhibits the best underwater adhesive performance, and the bond strength can achieve 2.41 MPa. Meanwhile, this adhesive exhibits excellent stability, whether in tap water or saltwater, whether under changed temperature (25–90 °C) or prolonged time (~180 days) conditions, which is of great importance in practical applications. And this adhesive will not cause any damage to substrates or leave hard-to-remove residues after bonding, which can be removed through swelling and erasing. And the finally successful leak repair of underwater metal cans and water pipes demonstrates the potential application of this adhesive in underwater and humid environments.

2 Experimental

2.1 Materials

Aluminum nitrate hexahydrate ($\text{Al}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, AR), anhydrous sodium carbonate (Na_2CO_3 , AR), oleic acid (OA, 90%), oleyl amine (90%+), ethanol (AR), and cyclohexane (AR) were purchased from Shanghai Adamas Co., Ltd. Perfluorodecyltriethoxysilane and

methyltrimethoxysilane were used as modifiers without further purification. All reagents were used directly as received unless otherwise stated.

2.2 Synthesis of CN ASNPs

First, 4 mL of oleic acid was added to 16 mL of anhydrous ethanol and stirred thoroughly until homogeneous. Then 1 mL of oleylamine was introduced, and the mixture was continuously stirred for 15 min until a yellow transparent solution was formed. Subsequently, 7 mL of 0.25 M anhydrous sodium carbonate solution was added, followed by stirring for 5 min. After that, 7 mL of 0.25 M aluminum nitrate solution was added to the above mixture, which was then sealed and stirred for 40 min. The resulting suspension was transferred to a centrifuge tube and centrifuged at 8000 r/min for 5 min. The supernatant was discarded, and a pale yellow-white colloidal viscoelastic substance was obtained. The product was dispersed in cyclohexane, and an equal volume of ethanol was added for thorough shaking and washing. The mixture was centrifuged again at 8000 r/min for 5 min, and the washing process was repeated twice. The final product was named CN ASNPs.

2.3 Synthesis of CN ASNPs-1 and CN ASNPs-2 adhesives

The preliminary synthesis steps of CN ASNPs-1 and CN ASNPs-2 were identical to those of CN ASNPs adhesive. Briefly, 4 mL of oleic acid was mixed with 16 mL of anhydrous ethanol, followed by the addition of 1 mL of oleyl amine and stirring for 15 min. After sequential addition of 7 mL of 0.25 M anhydrous sodium carbonate solution and 7 mL of 0.25 M aluminum nitrate solution, the mixture was sealed and stirred for 40 min. The suspension was centrifuged at 8000 r/min for 5 min, and the supernatant was discarded. The obtained viscoelastic substance was dispersed in cyclohexane, washed with an equal volume of ethanol, and centrifuged at 8000 r/min for 5 min. The product was then redispersed in 18 mL of cyclohexane, and an equal volume of mixed solution (modifier:ethanol = $x:y$, v/v, and the optimal ratio was determined to be 1:1) was added. For CN ASNPs-1, the modifier was FDETS; for CN ASNPs-2, MTMS was used. After thorough shaking and centrifugation at 8000 r/min for 5 min, white viscoelastic modified products were obtained and named CN ASNPs-1 and CN ASNPs-2, respectively.

3 Results and discussion

CN ASNPs were synthesized (Fig. 1(a)) via a stirring reaction under room-temperature and atmospheric pressure and showed good gelation performance. This method is not only simple but also enables direct large-scale production (Fig. 1(b)). The CN ASNPs can be well dispersed in non-polar organic solvents, such as cyclohexane, and tend to aggregate into a 3D network structure (Fig. 1(c)). And the obvious Tyndall effect indicated that a stable colloidal system formed (the inset in the bottom-left corner of Fig. 1(c)). After a period of standing still, the freestanding organogel can form (Fig. 1(d)). The scanning electron microscope (SEM) of aerogel obtained by freeze-drying (Fig. S1 in the Electronic Supplementary Material (ESM)) showed that non-uniformly sized bubbly pores and a robust 3D network structure coexist, demonstrating the excellent locking ability for organic solvents. Energy-dispersive X-ray spectroscopy (EDS) analysis (Fig. S2 in the ESM) suggested that CN ASNPs contained aluminum, carbon and

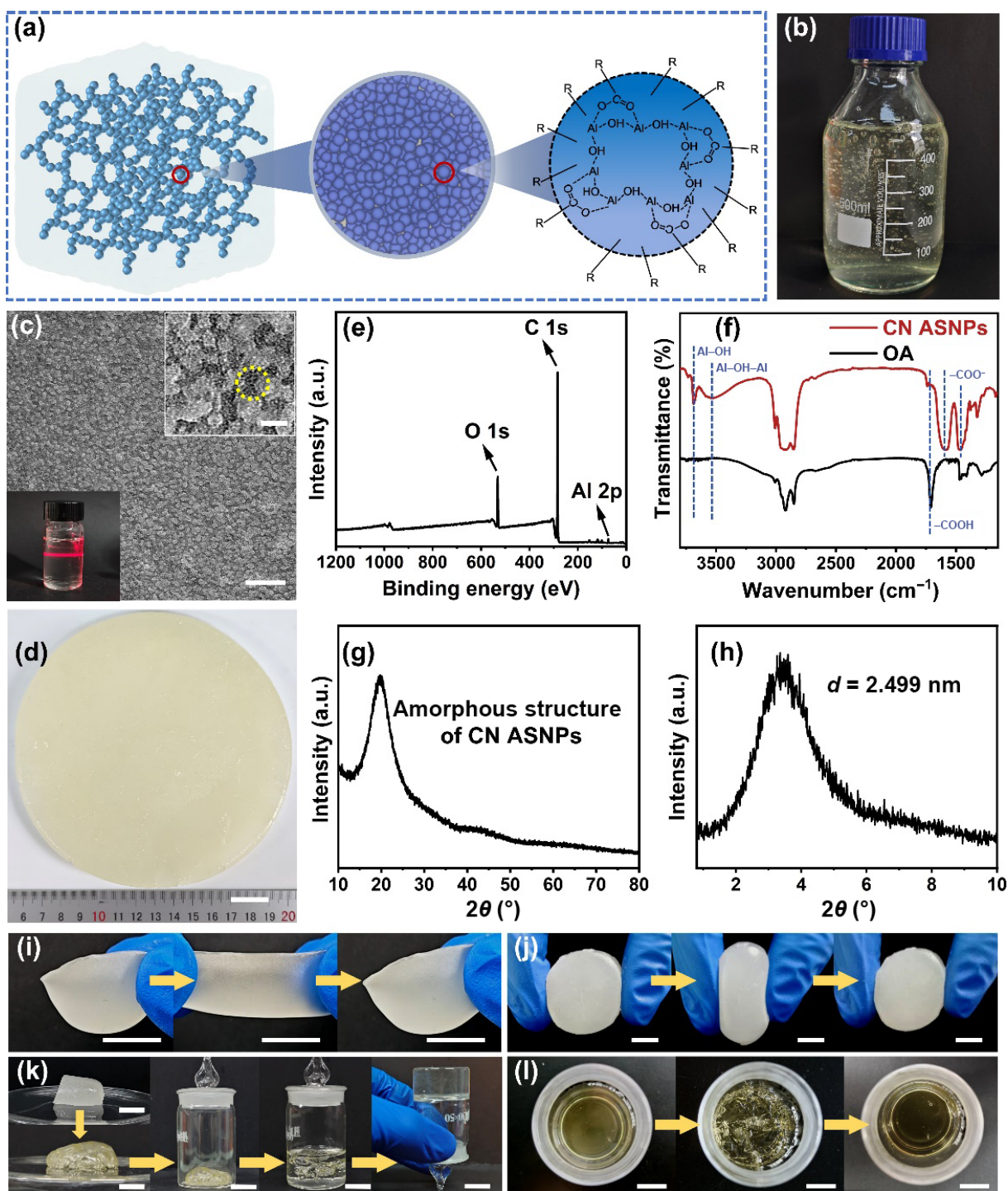


Figure 1 Characterizations of CN ASNPs and gels. (a) Schematic diagram of CN ASNPs-based organogel. (b) Mass-producible photograph of CN ASNPs. (c) TEM images of the CN ASNPs. Scale bar is 50 nm. The inset in the bottom-left corner shows the CN ASNPs cyclohexane dispersion and its Tyndall effect. The inset in the top-right corner shows the high-magnification TEM image of the CN ASNPs. Scale bar is 10 nm. (d) Photograph of an CN ASNPs cyclohexane organogel with a diameter of ~ 14 cm and thickness of ~ 1.5 cm. Scale bar is 2 cm. (e) XPS spectrum of CN ASNPs. (f) FTIR spectra of CN ASNPs. (g) XRD pattern of CN ASNPs. (h) SAXRD pattern of CN ASNPs. (i) and (j) Elasticity demonstration of CN ASNPs organogels. Scale bars are 1 cm. (k) Reversible conversion of the dried gel and wet gel. Scale bars are 1 cm. (l) Self-healing property of CN ASNPs organogel. Scale bars are 1 cm.

oxygen, and these elements were uniformly distributed. Furthermore, the X-ray photoelectron spectroscopy (XPS) (Fig. 1(e) and Fig. S3 and Table S1 in the ESM) results also demonstrated this result. As shown in the Fourier transform infrared (FTIR) spectra (Fig. 1(f)), there is no obvious peak of carboxyl groups in the CN ASNPs, and two characteristic peaks of carboxylate groups appear at 1584 and

1463 cm^{-1} [54]. Meanwhile, it is demonstrated that the carboxylate groups adopt the bridging bidentate coordination mode, connecting two adjacent Al^{3+} centers. The wide peak of 3200–3600 cm^{-1} and sharp peak of 3680 cm^{-1} belong to hydroxy bridges (Al–OH–Al) and terminal hydroxyl (Al–OH), respectively. There was only a single peak in the X-ray diffraction (XRD)

results (Fig. 1(g)), suggesting the amorphous structure of CN ASNPs. There is an obvious peak in the small-angle X-ray diffraction (SAXRD) result (Fig. 1(h)), revealing that the CN ASNPs formed an ordered packing structure, with an interparticle spacing of approximately 2.499 nm. Combining the above results, the structure of the CN ASNPs is illustrated in Fig. 1(a). The aluminum centers are polymerized via olation (hydroxy bridging, Al-OH-Al), forming the inorganic framework, and the carboxylate ligands adopt the bridging bidentate coordination mode, connecting adjacent aluminum sites and further crosslinking the network.

As shown in Fig. S4 in the ESM, the viscosity of CN ASNPs cyclohexane dispersions increased significantly with rising mass fraction of CN ASNPs, forming a gel at the critical gel concentration (10.95%) that exhibits excellent adhesion (inset in Fig. S4 in the ESM, adhering well to nitrile gloves). Rheological tests on samples with mass fractions of 4.01%, 9.39%, 18.30%, and 23.92% (Fig. S5 in the ESM) revealed that increased CN ASNPs content significantly enhanced the G' and G'' , indicating a denser 3D network, strengthened cohesion, and improved viscous dissipation. As shown in Figs. 1(i) and 1(j), the freestanding CN ASNPs organogel with outstanding elasticity forms, and whether compressed or stretched, it can restore its initial state. Notably, the dried gel and organogel can be converted to each other by swelling and drying. Even when the dried gel was ground into powder, an organogel can form again through redispersion and standing (Fig. 1(k) and Fig. S6 in the ESM), demonstrating excellent redispersibility and reconstitution ability of CN ASNPs. In addition, the CN ASNPs organogel had self-healing property. As shown in Fig. 1(l), an organogel was crushed, and after a few moments, the gel can recover as before. The gelation performance of CN ASNPs was also tested using various organic solvents (cyclohexane, toluene, chloroform, hexane, n-octane, oleyl alcohol, octadecene, Fig. S7 in the ESM), and critical gel concentrations (CGCs) were 10.95%, 7.66%, 6.96%, 11.81%, 9.47%, 20.35%, and 13.50% (Table S2 in the

ESM), respectively. Moreover, these organogels exhibited freeze-resistance, remaining intact after being immersed in liquid nitrogen for 30 min (Fig. S8 in the ESM). The mechanical tests showed that regardless of the solvent, higher CN ASNPs mass fractions lead to increased compressive stress at the same strain (Fig. S9 in the ESM), indicating the higher the mass fraction of CN ASNPs, the denser the internal 3D network and the stronger the skeleton structure [55]. At similar weight fractions, the octadecene- and oleyl alcohol-based gels exhibited the largest strain but relatively low tensile stress, while the hexane- and cyclohexane-based gels displayed the highest tensile stress but limited strain. The toluene-, chloroform-, and n-octane-based gels fell in between, with a more balanced combination of tensile stress and strain (Fig. S10 in the ESM).

To investigate polymerization process of CN ASNPs, a series of CN ASNPs under different reaction times is prepared. As shown in Fig. 2(a), gel permeation chromatography (GPC) was performed to determine the molecular weight changes of the products. When the reaction time was 30 s, the weight-average molecular weight (M_w) of the product was only 1620 g/mol. As the reaction time increased, the molecular weight gradually increased. At a reaction time of 40 min, the M_w reached 180,748 g/mol. It is proved that as the reaction time prolongs, the degree of polymerization of the CN ASNPs increases. As shown in Fig. 2(b), the cyclohexane dispersions with a mass fraction of 4% were prepared with these CN ASNPs. First, the viscosity of these dispersions was quantitatively measured using a viscometer. When the reaction time was 1 min, the viscosity was only 16.7 mPa·s. As the reaction time increased, the viscosity gradually increased. And when the reaction time was 40 min, the viscosity had essentially stabilized. As shown in Fig. 2(c), after being subjected to the same degree of shaking, the number of bubbles in the cyclohexane dispersion gradually increased with prolonged reaction time, macroscopically indicating an increase in the viscosity and the formation of denser crosslinking network structures. As shown in Fig. 2(d), SAXRD was performed to investigate the variation of spacing between CN

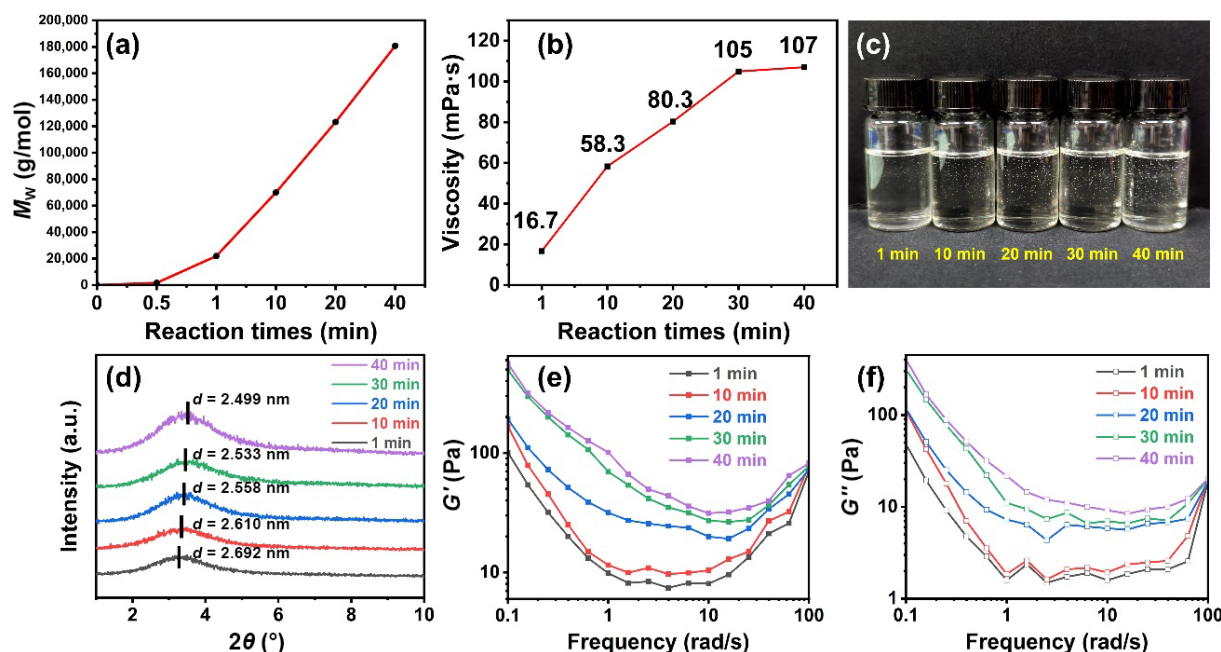


Figure 2 Characterizations of CN ASNPs under different reaction times. (a) Molecular weight of CN ASNPs under different reaction times. (b) Viscosity of CN ASNPs cyclohexane dispersions under different reaction times. (c) Photo of bubble situation under different reaction times. (d) The SAXRD pattern of CN ASNPs under different reaction times. (e) and (f) Rheological tests of CN ASNPs.

ASNP. The calculated spacings of CN ASNPs obtained at 1, 10, 20, 30, and 40 min were 2.692, 2.610, 2.558, 2.533, and 2.499 nm, respectively. With increasing reaction time, the spacing gradually decreased, indicating CN ASNPs are arranged more tightly. The above results demonstrate that the increase in polymerization degree leads to the increase in viscosity and density of crosslinking network of ASNPs. As shown in Figs. 2(e) and 2(f), rheological tests were conducted on the 4 wt.% products obtained at different reaction times. With increasing reaction time, both the storage modulus (G') and loss modulus (G'') gradually increased, indicating a simultaneous enhancement in the internal structural strength and viscous dissipation capacity of the products. When the reaction time reached 30 min, both moduli reached relatively high levels and tended to stabilize, confirming that 40 min of reaction time ensures good structural strength and gel performance of the product.

Because the CN ASNPs organogel exhibited high viscosity and good mechanical properties, its adhesive performance was further investigated. As shown in Fig. S11 in the ESM, with the increase of reaction time, the adhesive strength gradually increased, and when the reaction time reached 40 min, the improvement of adhesive strength tended to be stable, reaching 2.33 MPa. This indicates that the higher the degree of polymerization of ASNPs, the stronger the cohesion of CN ASNPs, thereby improving the adhesive strength. The addition amount of OA also had a great influence on the adhesive strength. As shown in Fig. S12 in the ESM, it is found that

when 4 mL OA was added in the reaction, the adhesive performance was the best, so this addition amount was adopted for subsequent tests. In addition to adhesive performance in air (Figs. S13 and S14 in the ESM), more importantly, the CN ASNPs organogel also demonstrated outstanding underwater adhesive performance. This adhesive could directly bond stainless steel weights underwater and immediately lift a certain load, and it could be maintained for at least 180 days (Figs. 3(a)–3(c)). The adhesive exhibited different adhesive performances on different substrate materials, and for stainless steel, it is the best. As shown in Fig. 3(d), the underwater adhesive strengths of stainless steel, aluminum, copper, ceramics and PA66 were 1.83, 1.22, 1.44, 0.878, and 0.548 MPa, respectively. Due to the hydration layer on the substrate surface, underwater and under-saltwater adhesive strengths were slightly inferior to that in air (Fig. 3(e)), but the performance was still good among reported underwater adhesives (Table S5 in the ESM). The curing time of the CN ASNPs organogel adhesive was also investigated. As shown in Fig. 3(f), the initial adhesive strength was only 0.343 MPa, which reached the maximum value after curing at room temperature and underwater environment for 24 h. The CN ASNPs organogel adhesive also exhibited excellent stability. As shown in Fig. 3(g), after being immersed in water for 90 days, the adhesive strength remained stable. And as shown in Fig. 3(h), after bonding underwater at 60 and 90 °C, the adhesive strengths only slightly decreased from that at room temperature.

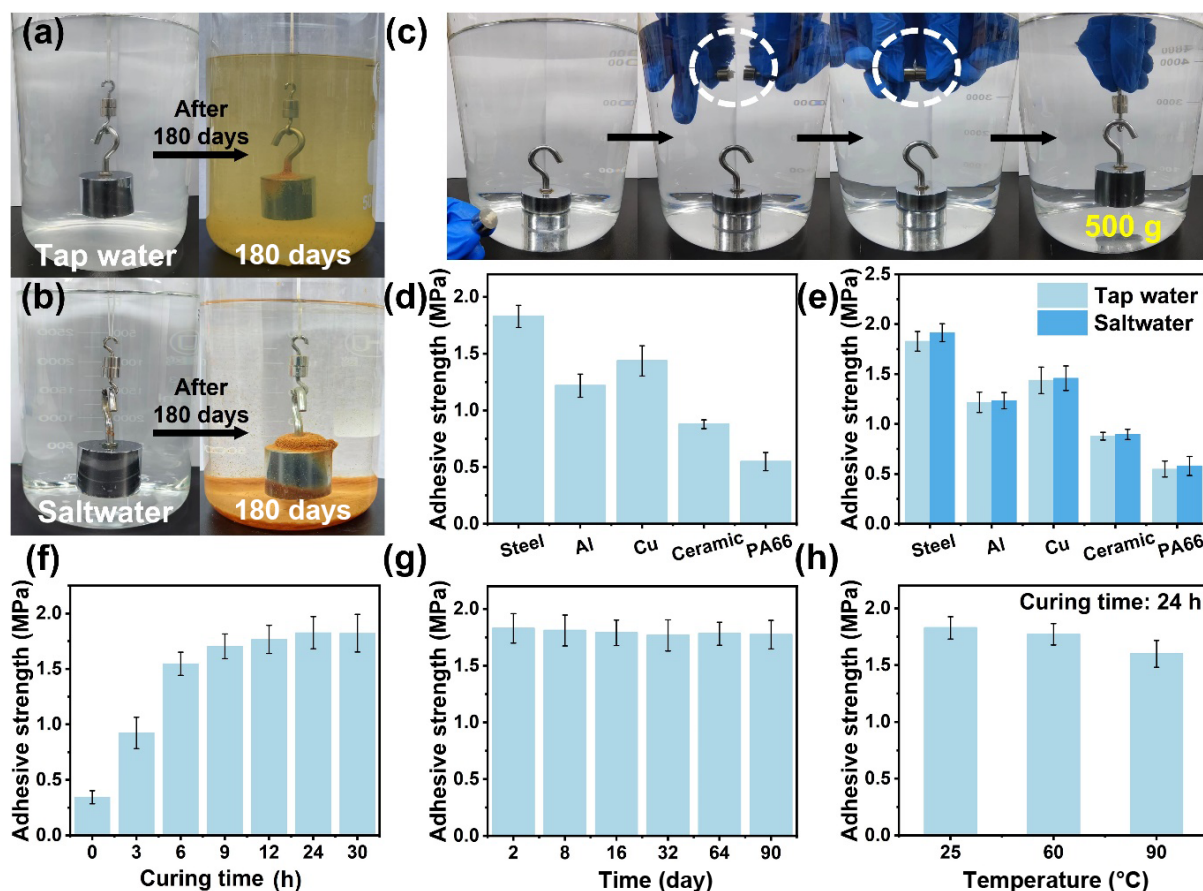


Figure 3 Underwater adhesive performance of CN ASNPs organogel adhesive. Comparison of weights bonded by CN ASNPs organogel adhesive immersed in (a) tap water and (b) 3.5% (mass fraction) NaCl saltwater after 180 days. (c) Underwater adhesion process with CN ASNPs organogel adhesive. (d) Adhesive strength of CN ASNPs organogel adhesive on diverse substrates. (e) Comparison of adhesive strength of CN ASNPs organogel adhesive on diverse substrates under tap water and 3.5% (mass fraction) NaCl saltwater. (f) Adhesive strength of CN ASNPs organogel adhesive on steel substrates after different curing time. (g) Long-term adhesion stability of CN ASNPs organogel adhesive on steel substrate. (h) Adhesive strength of CN ASNPs organogel adhesive cured at different temperatures.

Additionally, as shown in Fig. S15 in the ESM, when the bonded weights were immersed in cyclohexane solvent, the cured CN ASNPs gradually swelled with increasing immersion time, and the adhesion was completely released after 4 h, allowing for complete removal with tissue paper. This adhesive will not cause any damage to substrates or leave hard-to-remove residues after bonding.

Material hydrophobicity is a key factor affecting underwater adhesive performance [56]. Excellent hydrophobicity can minimize material dissolution and swelling, ensuring stability in humid environments [57]. To further improve the underwater adhesive performance of the CN ASNPs organogel adhesive, a simple and convenient hydrophobic modification strategy was adopted by introducing hydrophobic siloxane small molecules into CN ASNPs. Compared with chemical modification, this method is simple to operate efficiently and exhibits remarkable modification effects. FDETS and MTMS (Fig. S16 in the ESM) were selected as modifiers to prepare modified adhesives CN ASNPs-1 and CN ASNPs-2, respectively. XPS results (Figs. S17–S20 and Tables S3 and S4 in the ESM) demonstrated the presence of FDETS or MTMS among CN ASNPs. EDS analysis of liquid nitrogen-frozen CN ASNPs-1 and CN ASNPs-2 (Figs. S21 and S22 in the ESM) further revealed uniform distribution of FDETS or MTMS,

confirming successful grafting of siloxane modifiers onto CN ASNPs.

In the FTIR results of CN ASNPs-1, the peak at 1192 cm^{-1} was attributed to the Si–O–C bonds of FDETS, confirming the successful introduction of FDETS (Fig. 4(a)). Compared with FDETS, the peaks attributed to Si–O–C bonds were split from 1100 into 1080 cm^{-1} , assigned to Si–O–Si bonds, and 890 cm^{-1} , assigned to Si–OH bonds (Fig. 4(b)). This change could be attributed to hydrolysis of MTMS within ASNPs-2 [58, 59]. Meanwhile, partial hydrolysis significantly increased the number of hydroxyl groups within ASNPs-2, as verified by the strengthening of the peaks at $3600\text{--}3200\text{ cm}^{-1}$. In addition, a shift in the peak ($3600\text{--}3200\text{ cm}^{-1}$) was attributed to the formation of hydrogen bonds between ASNPs and MTMS [60, 61]. A plenty of hydroxyl groups can not only form high-density hydrogen bonds to improve cohesion within the adhesive but also strengthen interactions between the adhesive and substrates. Moreover, the hydrolysis induced polycondensation (Si–O–Si) effectively enhanced cohesion, thereby helping to further improve the adhesion performance of the adhesive. Similar to the unmodified CN ASNPs aerogel, SEM images of CN ASNPs-1 and CN ASNPs-2 (Figs. 4(c) and 4(d)) aerogels showed robust 3D frameworks with non-uniform bubbly pores. And SAXRD results

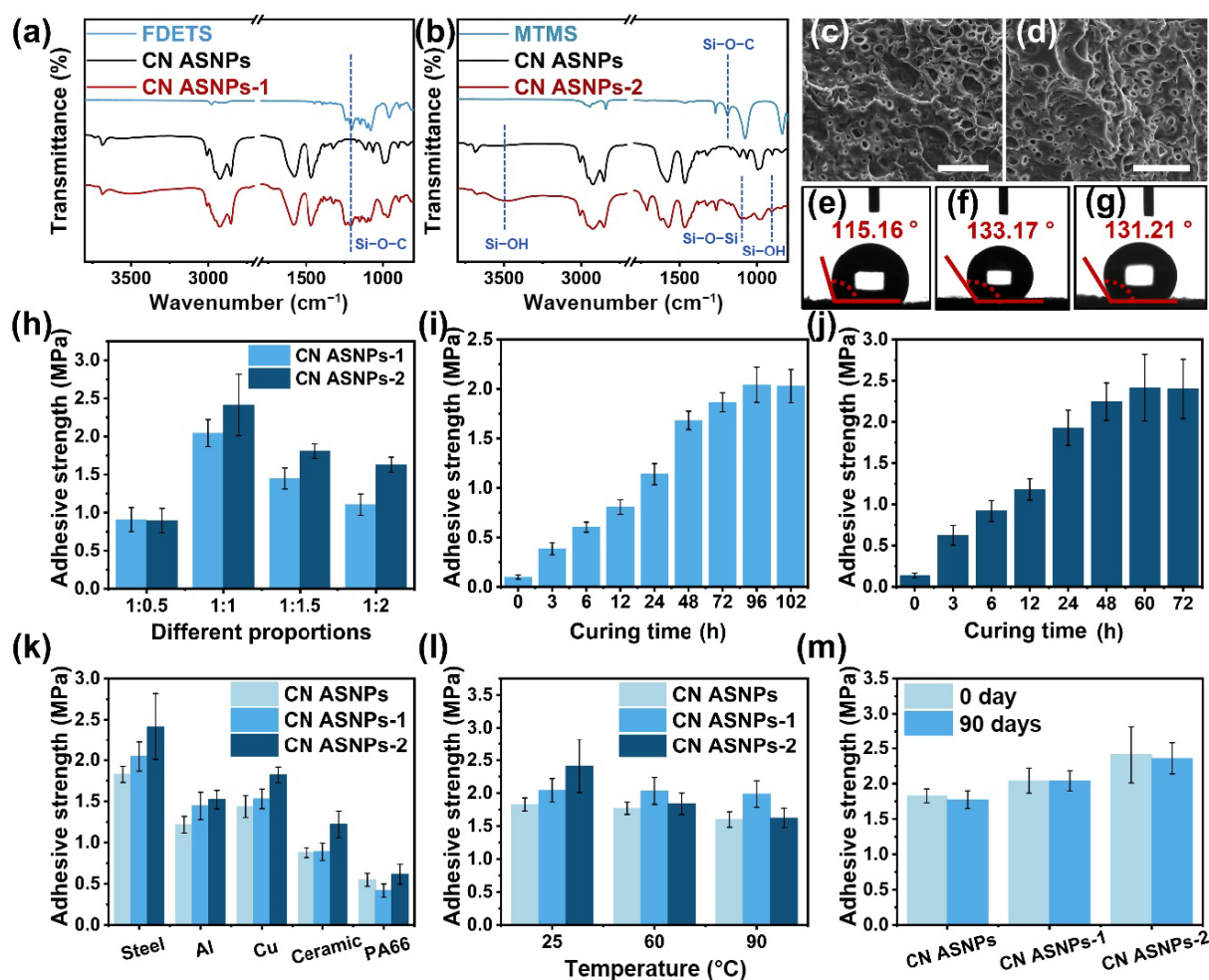


Figure 4 Characterization and the underwater adhesion performance of CN ASNPs after modification. FTIR spectra of (a) CN ASNPs-1 and (b) CN ASNPs-2. SEM images of (c) CN ASNPs-1 and (d) CN ASNPs-2. The water contact angles of (e) ASNPs, (f) CN ASNPs-1, and (g) CN ASNPs-2. (h) Adhesive strength of CN ASNPs modified by different proportions of hydrophobic solvents. Adhesive strength of (i) CN ASNPs-1 and (j) CN ASNPs-2 on steel substrates after different curing times. Comparison of adhesive strength of ASNPs, CN ASNPs-1 and CN ASNPs-2 (k) on various substrates, (l) cured at different temperatures and (m) immersed in tap water after 90 days.

(Fig. S23 in the ESM) also indicated the ordered particle packing, and interparticle spacings were ~ 2.414 nm (CN ASNPs-1) and ~ 2.355 nm (CN ASNPs-2), respectively. The above results showed that the modification had no obvious damage to the structure of ASNPs organogel. Contact angle (CA) tests (Figs. 4(e)–4(g)) demonstrated that the initial CA of unmodified CN ASNPs (115.16°) increased to 133.17° (CN ASNPs-1) and 131.21° (CN ASNPs-2). This increase facilitated disruption of the substrate surface hydration layer and thereby promoted interfacial adhesion between the adhesive and substrate.

Further, the underwater adhesive performance of CN ASNPs-1 and CN ASNPs-2 was tested. First, the optimal addition amount of modifier was investigated, the modifier was introduced to the CN ASNPs organogel adhesive by using ethanol containing this modifier as a flocculant (see the Methods section in the ESM for details). And when the volume ratio of ethanol to modifier reached 1:1, the adhesive performance was the best (Fig. 4(h)). This ratio was adopted to do the subsequent tests. Then, curing time was also an affecting factor for the adhesive performance. Whether CN ASNPs-1 or CN ASNPs-2 exhibited increased adhesive strength with the curing time increasing (Figs. 4(i) and 4(j)). As shown in Fig. 4(k), both CN ASNPs-1 and CN ASNPs-2 showed better adhesive performance than the unmodified CN ASNPs adhesive on various substrates, while CN ASNPs-2 was better than CN ASNPs-1. This phenomenon might be attributed to the presence of perfluorinated chains within CN ASNPs-1, which decreased the surface energy, inhibiting the surface spreading of the adhesive on the substrate surface, consequently resulting in lower adhesive strength than CN ASNPs-2 [62–64]. Although hydrophobicity was enhanced, it also brought about certain negative effects. The temperature stability of CN ASNPs-1 and CN ASNPs-2 was tested, as shown in Fig. 4(l). Compared to room temperature, after bonding underwater at 60 and 90 $^\circ\text{C}$, the adhesive strengths of CN ASNPs-1 remained stable, in contrast, there was a slight decrease in the adhesive strengths of CN ASNPs-2 due to that the hydrogen bonds generated by MTMS exhibit instability upon heating. The long-term stability tests of CN ASNPs, CN ASNPs-1, and ASNPs-2 were performed. As shown in Fig. 4(m), the adhesive performance remained stable after 90 days of immersion in water at room temperature.

The underwater adhesion mechanism of CN ASNPs series adhesives was investigated. Taking a stainless-steel substrate as the model, the failure modes of the CN ASNPs series adhesives were studied by evaluating their tensile fracture surfaces. As shown in Figs. 5(a)–5(f), cohesive failure occurs partially within the adhesive and partially at the surface of the bonded substrate, and was observed on the fracture surfaces of ASNPs, CN ASNPs-1, and CN ASNPs-2 adhesives, indicating a mixed failure mode of cohesive failure and interfacial failure [65–70]. This mixed failure mode provided effective support for adhesive strength, and the formation of this mode depends not only on the inherent properties of the adhesive but also on the interactions between the adhesive and the substrate. Additionally, many tear-like morphologies were observed on the fracture surfaces of CN ASNPs-1 and CN ASNPs-2 adhesives, indicating a certain degree of layered fracture in the modified adhesives. It can be ascribed to a relative balance between cohesion and adhesion in the CN ASNPs-1 and CN ASNPs-2 adhesives, but under this balanced state, the CN ASNPs-1 and CN ASNPs-2 adhesives withstood higher fracture tensile force over CN ASNPs organogel adhesive, therefore, during the tensile fracture process, significantly improved stress dissipation.

As shown in Fig. 5(g), when modified CN ASNPs organogel adhesives were adhered underwater to the substrate, the enhanced hydrophobicity led to the more efficient destruction of the hydration layer and the exclusion of a large number of water molecules from the substrate surface, maximizing the adhesion area, thereby promoting close contact between the adhesives and the substrate. After close contact with the substrate, the adhesive could form micro- to nanoscale mechanical interlocking with the micro-rough structure on the substrate surface, which not only enhanced adhesion performance but also strengthened surface interactions, including van der Waals forces, metal coordination, electrostatic interactions, and hydrogen bonds. Moreover, the hydrogen bonding interactions within the modified adhesive and the polycondensation of silane molecules provided important support for high cohesion.

Notably, although CN ASNPs-1 had stronger hydrophobicity, the underwater adhesion performance of CN ASNPs-2 was improved more significantly, indicating that hydrophobicity was not the only effect factor for preparing high-performance underwater adhesives. The presence of perfluoro chains in CN ASNPs-1 enhanced the ability to destroy the hydration layer but inhibited the affinity between the adhesive and the substrate, resulting in limited improvement of adhesion performance. However, in CN ASNPs-2, moderate hydrophobicity effectively disrupted the hydration layer, partial hydrolysis generated more hydroxyl groups to enhance interfacial adhesion, and the polycondensation of silane molecules collectively achieved a significant improvement in adhesive strength. In summary, hydrophobicity, interfacial adhesion, and structural cohesion were all critically important in the design of high-performance underwater adhesives.

Liquid leakage occurring underwater is a very thorny problem, so underwater leak sealing measures have been widely studied by people. Based on the above, the CN ASNPs series adhesives demonstrated excellent underwater adhesion performance, showing great potential for underwater plugging. As illustrated in Figs. 5(h) and 5(i), the adhesives successfully achieved leak sealing of stainless steel and aluminum tanks with leaking liquids in an underwater environment. For evaluating the stability of leak sealing, Rhodamine B was employed to stain the leaking liquid, and water samples were collected at different time intervals for ultraviolet (UV) absorption measurements (Fig. S24 in the ESM). The results demonstrated that after the three adhesives were applied for leak sealing and stored for 60 days, there was no significant variation in the intensity of the characteristic absorption peak of Rhodamine B around 550 nm, confirming the long-term stability of their leak sealing performance. In addition, a leak sealing test was conducted on a 1.5 m-long iron pipe filled with water. Following pressure-assisted curing, the iron pipe was successfully sealed, and the adhesive effect remained stable after 60 days of storage (Fig. 5(j)), which further verified the application value of CN ASNPs series adhesives in practical engineering scenarios.

4 Conclusion

In this study, CN ASNPs was successfully synthesized via a facile stirring reaction at room-temperature and atmospheric pressure, enabling large-scale production and laying a solid foundation for practical applications. Mechanistic investigations revealed that the aluminum centers polymerize through ololation to form an inorganic

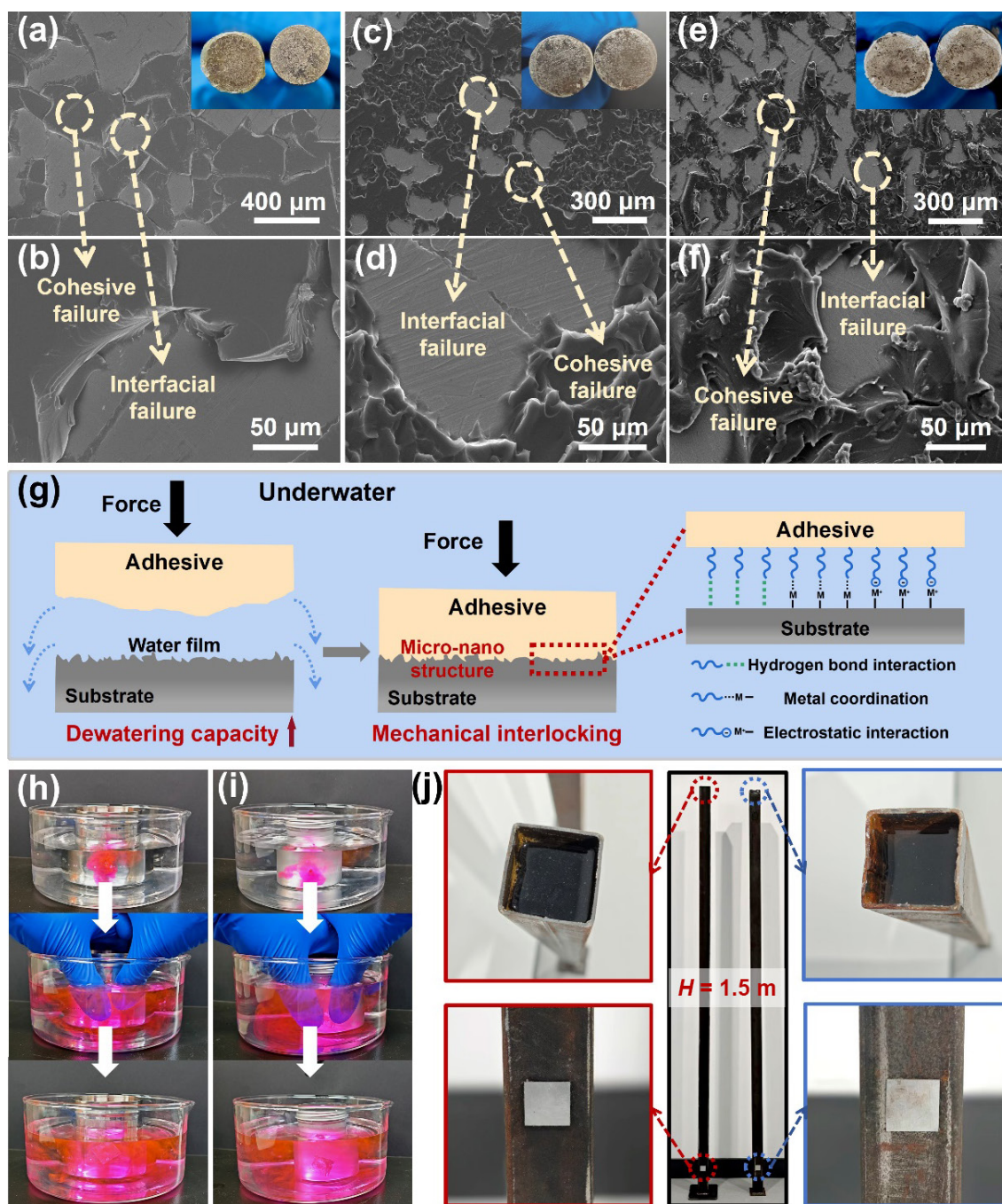


Figure 5 The adhesion mechanism and applications in leaking stoppage. (a)–(f) SEM images of ((a) and (b)) CN ASNPs, ((c) and (d)) CN ASNPs-1, and ((e) and (f)) CN ASNPs-2 on two separated adherends after the bonding was broken (the insets were photographs of the fractured adhesive surfaces). (g) Schematic diagram of adhesion mechanism. Photographs of CN ASNPs-2 directly plugging leaks on (h) iron cans and (i) aluminum cans underwater. (j) Photographs of a 1.5-meter-long iron pipe filled with water leak-plugged by CN ASNPs (left) or CN ASNPs-2 (right) after 60 days.

framework, while carboxylate ligands adopt a bridging bidentate coordination mode to connect adjacent aluminum sites, thereby constructing a highly crosslinked network. With increasing reaction time, the degree of polymerization of CN ASNPs significantly increased, leading to the formation of a denser three-dimensional network structure. This directly endowed the organogel with excellent viscoelastic properties and cohesive strength. The CN ASNPs organogel adhesive exhibits outstanding underwater adhesion performance. This remarkable performance arises from the synergistic effect of hydration layer disruption, micro-nano

mechanical interlocking, a high degree of polymerization, and abundant intermolecular forces. After modification by FDETS and MTMS, the hydrophobicity and cohesion were further enhanced, resulting in improved adhesive strength and interfacial binding energy. The adhesive demonstrates broad applicability to various substrates, including steel, aluminum, copper, ceramics, and PA66 and the underwater adhesive strength on steel can achieve ~ 2.41 MPa. Moreover, it maintains stable adhesion under harsh conditions, such as immersion in tap water or saltwater for 180 days, and temperature variations from 25 to 90 °C. Importantly,

the adhesive can be easily removed by swelling without causing any damage to the substrates or leaving stubborn residues, highlighting its excellent engineering friendliness. In practical application tests, the CN ASNs organogel adhesive successfully achieved rapid leak sealing of underwater metal cans and water pipes, remaining leak-free for at least 60 days, thus demonstrating its reliability in emergency repair and long-term sealing scenarios. Looking forward, this sub-nanoparticle crosslinking network system holds great promise for broader applications in extreme engineering environments, such as deep-sea equipment maintenance and underwater structural repair. Systematic evaluation of its long-term durability and environmental adaptability will be crucial to accelerating its industrial adoption.

Electronic Supplementary Material: Supplementary material (experimental procedure, characterizations; tests of samples) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909043>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work. Author Xun Wang is the associate editor of this journal, but he is not involved in the peer-review or decision of this article.

Author contribution statement

S. G. Q., Y. F. Q., and J. X. L.: Data curation, project administration, validation, writing manuscript, experimental design. S. L.: validation. X. W.: Research direction. S. M. Z.: Project supervision, funding acquisition. All the authors have approved the final manuscript.

Use of AI statement

None.

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