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Is porous boron nitride suited for water treatment? – a critical perspective on the materials water stability

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Treatment of wastewater from organic and inorganic pollutants is an important process to secure access to clean drinking water in the future. A simple and cost-effective method to ensure the removal of trace contaminants is their adsorption with porous materials. Recent adsorption studies indicate that porous boron nitride (PBN) could remove pollutants like dyes, antibiotics, and metal ions very effectively due to its high surface area and partially ionic polar bond type, which greatly promotes interactions between the material and pollutants. Furthermore, these studies suggest good regeneration and high cycle stability of the PBNs. However, research on the water stability of PBN depicts extremely low hydrolytic stability and rapid decomposition in the presence of water. Here, multiple studies suggest that highly porous and defect rich BN is unsuited for applications in water. In order to understand this contradiction, a detailed literature review on the application of PBN in wastewater treatment and the materials hydrolytic stability is performed. Key properties of PBN for water treatment are identified and discussed critically in regard to its effect on stability. This correlation reveals clear gaps and new questions in the current research and provides new suggestions for future investigations.

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1. Introduction

The constant growth in the human population poses increasing demands on our society to guarantee access to basic human needs worldwide. Universal healthcare, food supply, and access to clean drinking water are already not secure in less developed countries.[1] Resource scarcity in particular poses a major problem for the future. As only 2.5% of the global water can be deemed as fresh water sources, water treatment is an important process to ensure an accessible and safe water supply despite its rising demand.[2,3] Years of industrial development have greatly contaminated water sources with persistent pollutants such as heavy metal ions or organic molecules like dyes, antibiotics, microplastics, and per- and polyfluoroalkyl substances (PFAS).[4–9] These are usually highly resistant to most common water treatment methods but pose great threats to the environment and human health. Thus, these compounds have to be removed by incorporating additional treatment steps like ion exchange, membrane filtration, advanced oxidation, ozonation, coagulation, or adsorption.[10,11] Among these methods, adsorption is one of the most commonly used due to its simplicity, high selectivity, and cost-effectiveness.[11] A large number of different materials have already been studied as adsorbents in wastewater treatment, like activated carbons,[12,13] MXenes,[14,15] zeolites,[16,17] silicates,[18] metal and covalent organic frameworks,[19,20] or porous boron nitrides (PBNs).[21,22] Especially, PBN has gained increasing attention for wastewater treatment in recent years. Due to its high temperature stability, large specific surface area, chemical inertness, and dipolar bond type, the material depicts very high adsorptive loadings for dyes, antibiotics, and heavy

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metal ions, greatly surpassing previous adsorbents.[3,23] In addition, boron nitride (BN) can be employed in the photocatalytic degradation of organic pollutants, further broadening its scope.[24] Until now, multiple reviews on the application of different BN species in water remediation have been published. The studies examine critical research fields like summarizing recent advancements of BN in pollutant adsorption[25–28] and water treatment[24,29–31] or the modification of BN materials for those applications.[32–34] However, these review articles primarily focus on the outstanding performance of BN without addressing the materials limitations during water exposure in greater detail. Recent studies[35,36] suggest that PBNs could be unsuited or unusable for water treatment due to very low hydrolytic stability. This shortcoming can potentially limit its application in water as the material could not be used for prolonged exposure periods.[37] Furthermore, the decomposition of PBN would lead to the release of hazardous compounds in the water, questioning the true use of PBN to remove pollutants. Here, a critical evaluation of the practical application of PBN for water purification is lacking in the literature. Therefore, this review article aims to look into the water treatment performance of BN in a different way by discussing its performance in relation to its water stability. To achieve this, at first, an introduction on the properties and synthesis methods of PBN is given, followed by a clear overview of its application in water purification. Here, a focus is set on the regeneration and cyclic stability of PBN, as well as known studies on the material properties post exposition. The last chapter covers a critical examination of the water stability of PBN with the aim to gain potential insights into how to improve the stability of PBNs in water treatment applications in future research and to identify research gaps in the literature that are so far unaddressed. Thereby, this article identifies multiple areas of research that must be addressed moving forward before PBN can be employed in water treatment.

2. Properties and structure of boron nitride

Boron nitride is formed by an equivalent number of alternating boron and nitrogen atoms that are arranged in different crystalline structures: cubic, rhombohedral, wurtzite-like, or hexagonal.[38,39] An overview of the different crystal structures is given in Fig. 1(a). Among those, hexagonal boron nitride (h-BN) is the most stable under standard conditions. In bulk (3D), the material depicts a layered structure. Each layer is formed by covalent sp^2-hybridized B–N rings. The individual layers are stacked ecliptically in an AA′A stacking order on top of each other.[40] Between the layers, weak van der Waals interactions arise with an interlayer spacing commonly between 0.330 to 0.333 nm.[41,42] This particular spatial arrangement

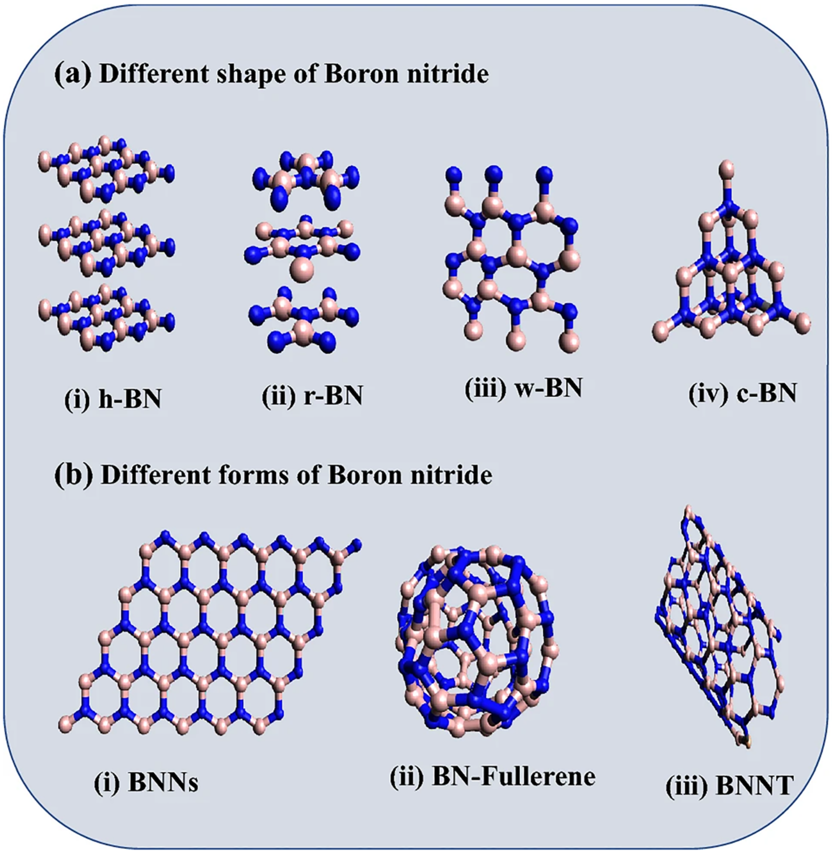


Fig. 1 Different (a) shapes and (b) forms of BN nanostructure materials. Reprinted from ref. 48. Reproduced with permission from Springer Nature. Copyright 2022 Springer Nature.

results from aromatic properties and strong polarization of BN due to the large discrepancy in electronegativity (EN) between the boron (EN = 2.04) and nitrogen (EN = 3.04) atoms, which leads to repulsive B ↔ B and N ↔ N or attractive B ↔ N interactions.[43] The large difference in EN results in a partially ionic bond type with "lopsided" density characteristics of the B–N bond.[44,45] Therefore, on the materials surface nitrogen atoms can act as Lewis base and boron atoms as Lewis acid centers.[46] Next to its bulk 3D-structure, h-BN can also be found in other dimensionalities like 2D boron nitride nanosheets (BNNS), 1D boron nitride nanotubes (BNNT), or 0D fullerene-like BN.[44,47] The different dimensionalities are depicted in Fig. 1(b).

In recent years, h-BN has been increasingly addressed in academic research due to its outstanding chemical and physical properties. Hexagonal BN in particular is interesting due to its very high thermal and chemical resistance. Kostoglou *et al.*[49] investigated the thermal stability of h-BN nanoplatelets in constant air flow using a thermogravimetric setup. Their experiments showed oxidation stability up to ~1000 °C. Afterwards, a mass increase of approximately 30% was observed by further increasing the temperature to 1200 °C, indicating the oxidation of h-BN to boron oxide (BO). In an inert or reducing atmosphere, h-BN depicts even higher stability up to 2000 °C.[50] Furthermore, h-BN is chemically inert and has low reactivity, making it exceptionally resistant towards most reaction conditions.[51] Crystalline h-BN depicts a large indirect band gap of ~5.9 eV. Thus, the material is often used as an insulation material for various electrochemical applications.[52]

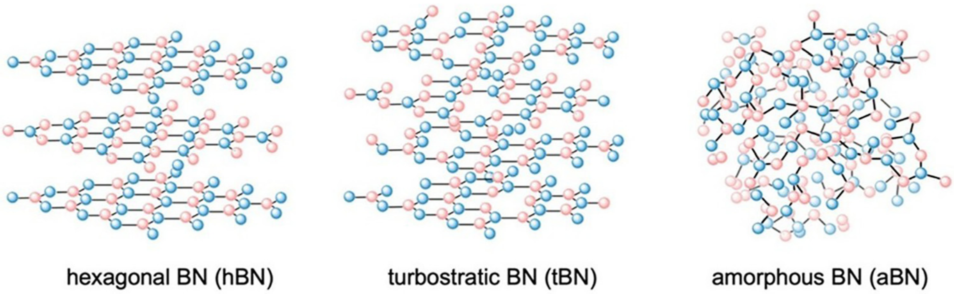
Despite these beneficial properties, there are also limitations to the use of h-BN. Due to its high crystallinity and tightly layered structure, h-BN usually has a very low specific surface area, limiting its adsorption and catalytic performance.[53] This greatly inhibits competitiveness with other porous materials in the aforementioned areas. To overcome this problem, defects can be generated in the crystal structure of h-BN to create gaps in the h-BN layers or to increase the interlayer stacking distance. This can either be accomplished by introducing new defects in the preformed h-BN structure with post-treatment methods like exfoliation,[54] or by directly synthesizing porous BN in template-free or template-assisted synthesis approaches.[55] The resulting less-ordered porous structures are no longer referred to as h-BN but rather as mesographitic, turbostratic, or amorphous BN depending on their crystallite size and interlayer spacing. According to Hubáček *et al.*,[56] a mean crystallite size above 60 nm is associated with h-BN, a mean crystallite size between 60 to 12 nm is counted to mesographitic boron nitride (m-BN), and mean crystallite sizes smaller than 12 nm belong to turbostratic boron nitride (t-BN). Thus, in the literature, m-BN and t-BN are referred to as a "semi-crystalline" material with a structure between amorphous BN and crystalline h-BN.[57] Nowadays, the distinction between m-BN and t-BN is rarely used, and porous BN is primarily referred to as amorphous or t-BN in the literature, depending on X-ray diffraction (XRD) data or interlayer spacing of the BN layers.

As shown in Fig. 2, t-BN is structurally similar to h-BN. However, instead of a clear stacking for t-BN, the BN layers are randomly twisted against each other. This results in the formation of small gaps in the structure, which increases the specific surface area while decreasing the crystallinity of the material. As a result of the loss in crystallinity, the reactivity of t-BN increases and thus its stability decreases. Kostoglou *et al.*[49] showed that there is a profound correlation between the oxidation stability of BN and its surface area. The group observed an exponential decrease in the materials oxidation temperature between approximately 800 to 1100 °C with increasing BET surface area. Prior research on the hydrolytic stability of BN also suggests that the reduction in crystallinity makes BN prone to hydrolysis.[58]

3. Synthesis of porous boron nitride

3.1. Top-down synthesis

Top-down synthesis methods refer to applications that introduce porosity in previously synthesized crystalline h-BN by exfoliation.[44,59] The exfoliation of h-BN to BNNS is primarily carried out either mechanically, by sonication in the liquid phase, or thermally. In mechanical exfoliation, generally, the h-BN material is ground in a ball mill, aiming to break larger crystallites into smaller fragments.[60] To prevent aggregation of the exfoliated sheets, usually solvents or surfactants are added before milling.[61–64] In sonication-assisted liquid phase exfoliation, h-BN is sonicated in a solvent with a surface tension close to the surface energy of the h-BN layers. Thereby, the solvent can overcome the interlayer stacking forces in h-BN and separate the layers from each other.[60] Zhu *et al.*[59] used thermal exfoliation of h-BN to synthesize porous BNNS. In their work,



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the authors heated h-BN first at 800 °C in air before immersing the hot material in liquid nitrogen. Using nitrogen sorption analysis, the group was able to show that with this method, the specific surface area of BN could be increased from 10 to 278 m g^{-1}.

3.2. Bottom-up synthesis

The bottom-up synthesis of PBN can be achieved by multiple approaches, like solvothermal reactions,[65–68] template-assisted methods,[69–73] or template-free methods.[74–76] Previous works have shown that solvothermal and template-assisted methods, despite higher preparative effort, usually result in lower specific surface areas than a template-free synthesis. Therefore, the template-free synthesis of porous BN is most often used in literature due to its low preparative effort, easy variability, and superior product properties.[75] Fig. 3 depicts the general approach of a template-free BN synthesis. In the first step, a boron and a nitrogen precursor are homogenized. Thereby, multiple parameters such as the precursor species, precursor ratio, and homogenization method can be varied. After that, the precursors are transferred to a pyrolysis furnace. The reaction conditions, such as heating rate, temperature, reaction time, atmosphere, and flow rate can be adjusted. Due to the high variability of the template-free synthesis, multiple studies on the effect of the different synthesis parameters have been published.

In a template-free approach, as boron sources, primarily boron oxide[36,77] or boric acid[74,78–80] are used due to their low cost. Nitrogen sources are much more varied and include compounds like urea,[77,81,82] melamine,[83–86] biuret,[75] dicyanamide,[74,78] ammonium chloride,[79] sucrose,[80] or hexamethylenetetramine.[87] Among those, urea and melamine are the most studied in the literature. Marchesini *et al.*[75] discussed the effect of using different nitrogen precursors on the properties of porous BN. In their work, they pyrolyzed boric acid and urea, biuret or melamine at a set molar ratio at 1050 °C for 3.5 h in nitrogen flow. With these experiments they were able to demonstrate that the chosen nitrogen precursor greatly changes the porosity development during the formation of BN. Nitrogen sorption analysis revealed that if melamine was used as precursor primarily micropores were formed. The usage of biuret or urea on the other hand resulted in the formation of a micro-mesoporous material with different pore ratios depending on the precursor. Marchesini *et al.* attributed these results to the different decomposition mechanisms and temperatures of the nitrogen precursors.

Another parameter greatly affecting the porosity of BN in a template-free synthesis is the precursor ratio.[81,83,85] In their work, Weng *et al.*[74] showed that varying the precursor ratio of dicyanamide (Dcy) to boric acid (BA) from 1 : 1 to 4 : 1 greatly influenced the specific surface area and total pore volume of the samples. As depicted in Fig. 4(a) and (b), the specific surface area and pore volume increase up to a ratio of 3 : 1 before declining. Örnek *et al.*[85] showed that changing the precursor ratio not only affects the specific surface area but also the crystallinity of BN. In their work, they varied the precursor ratio of melamine to boric acid from 1 : 1 to 1 : 6. Their studies revealed that with an increasing ratio of boric acid, the crystallinity of the synthesized BN increases.

In addition to the precursor species, the reaction conditions greatly influence the properties of PBN. The parameters with the greatest effect are the set temperature program and the reaction atmosphere. As given in Fig. 5, studies of L'Hermitte *et al.*[88] identified that the formation of PBN starts around 800 to 900 °C *via* a reaction of carbon nitride and oxidic boron species as intermediates. Other works suggest that the start of the BN formation occurs already at temperatures of 220 to 300 °C, with the formation of boron–oxygen–nitrogen intermediates.[39,82,89] Thus, it is evident that changes in the temperature program can greatly influence BN formation and the materials crystallization. Studies by Jähnichen *et al.*[82] showed that adding pre-heating steps during precursor decomposition and intermediate formation at 100 to 400 °C can change the specific surface area and pore ratio of the product significantly. Research on the final synthesis temperature identified that higher temperatures[86,90,91] and prolonged dwell times[57] result in a stronger crystallization of BN, leading to a loss in specific surface area after a certain point. As depicted in Fig. 4(c) and (d), Weng *et al.*[74] showed that an increase in the synthesis temperature from 800 to 1200 °C reduces the specific surface area and total pore volume of the synthesized PBN microsponges. This loss was attributed by the authors, to the ordering and crystallization of the BN phases with increasing temperature.

The template-free synthesis of PBN can be carried out in a vacuum[92] or under a constant flow of nitrogen[93] or ammonia.[87,94] In general, a synthesis in gas flow is preferred, as the gas can act as additional nitrogen precursor during the reaction. Research by Takagati *et al.*[94] suggests that changes in the flow rate affect the crystallinity of PBN. In their work, the authors showed that an

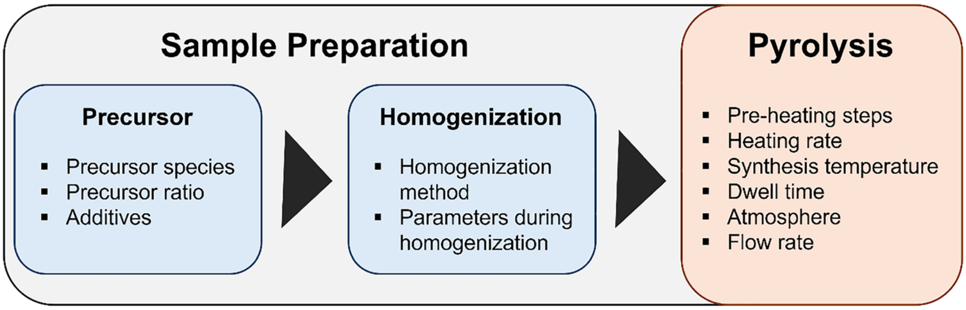

Fig. 3 General steps in the preparation and synthesis of boron nitride in a template-free synthesis approach.

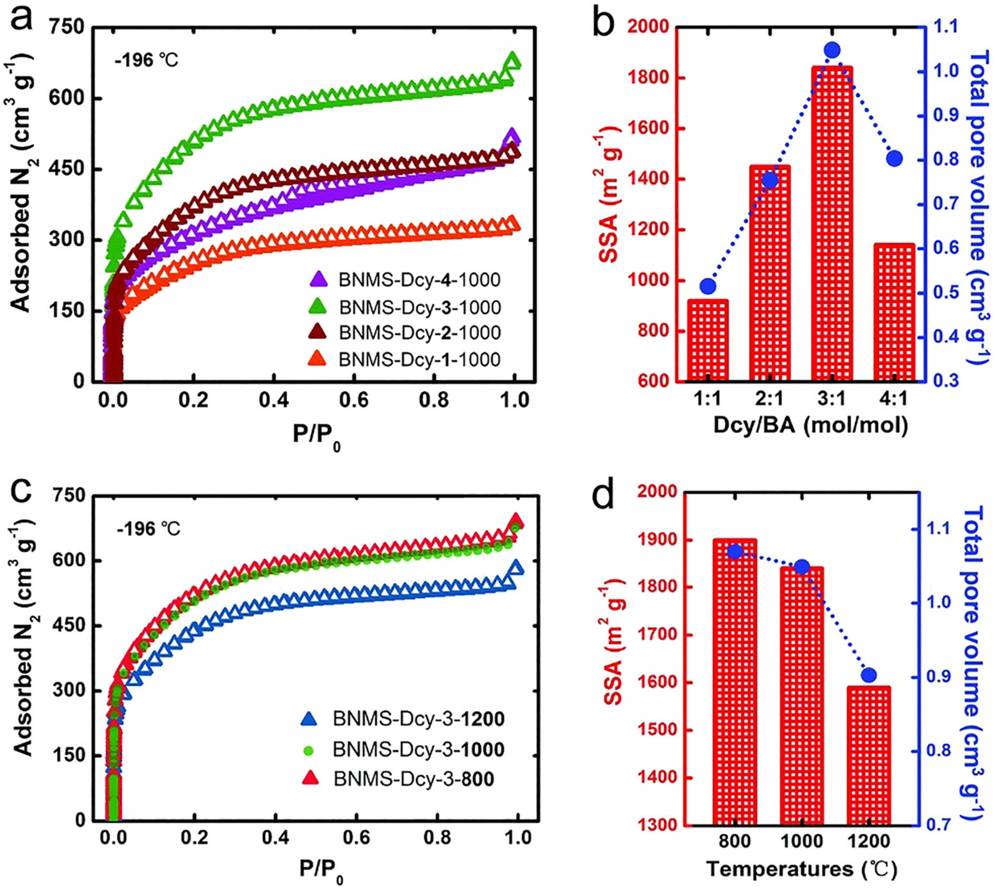

Fig. 4 (a) Nitrogen adsorption–desorption isotherms and (b) BET specific surface areas (SSAs) and total pore volumes of BN microsponges obtained with different Dcy/BA ratios at a fixed synthesis temperature of 1000 °C. (c) Nitrogen adsorption–desorption isotherms and (d) BET SSAs and total pore volumes of BNMSs obtained at different synthesis temperatures while keeping the ratio of Dcy/BA = 3 : 1. Reprinted with permission from ref. 74. Copyright 2013 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

increase in the ammonia flow during reaction from 50 to 150 mL min^{-1} reduced the crystallinity of the synthesized BN material. Later research of Shankar *et al.*[87] and Jähnichen *et al.*[93] depicted that changes in the flow rate also influence the amount of oxygen impurities and specific surface area of the product.

4. Stability of porous boron nitride in water treatment

4.1. Pollutant adsorption and degradation

Due to its high surface area, structural variability, Lewis acidic and basic adsorption sites, ''lopsided'' density characteristics, partially ionic bond type, and polar nature of PBN is a highly interesting adsorption material which has proven superior adsorption capacities to other porous adsorbents.[23,95–98] Li *et al.*[45] compared the adsorption performance of two PBN materials with activated carbon for tetracycline, methyl orange, and Congo red adsorption. As shown in Fig. 6(I), their investigation revealed much higher adsorption capacities of the BNs than the activated carbon for all three pollutants. The polarity of the covalent B–N bond and the aromaticity of BN greatly promote the adsorption of dyes and antibiotics by forming strong non-covalent dipole–dipole and π–π interactions, which lead to electrostatic attractions and π–π stacking on the BN surface.[99,100] The presence of B and N defect sites and functional –NH, B–O–B, and B–OH groups in PBN further enhances its interactions with hydrophilic groups in the molecules.[101–103] Liu *et al.*[104] studied the adsorption performance of highly porous BNNS on different antibiotics (tetracycline, chlortetracycline hydrochloride, ciprofloxacin, and norfloxacin) that are frequently detected in the environment and surface waters. The authors were able to identify up to ten times higher adsorption capacities for the antibiotics than generally reported in other studies. Furthermore, a strong pH dependence of the adsorption capacity was shown for ciprofloxacin and norfloxacin. For tetracycline and chlortetracycline hydrochloride, the effect was negligible. BN generally has its isoelectric point at low acidic pH values, as shown in Fig. 6(II), thus the surface charge of BN is primarily negative except in highly acidic conditions.[23,105] This greatly promotes the adsorption of zwitterionic or cationic pollutants and diminishes the adsorption of anionic pollutants at pH values greater than the isoelectric point of BN.[23,104,106] The organic pollutants, on the other hand, are more influenced by changes

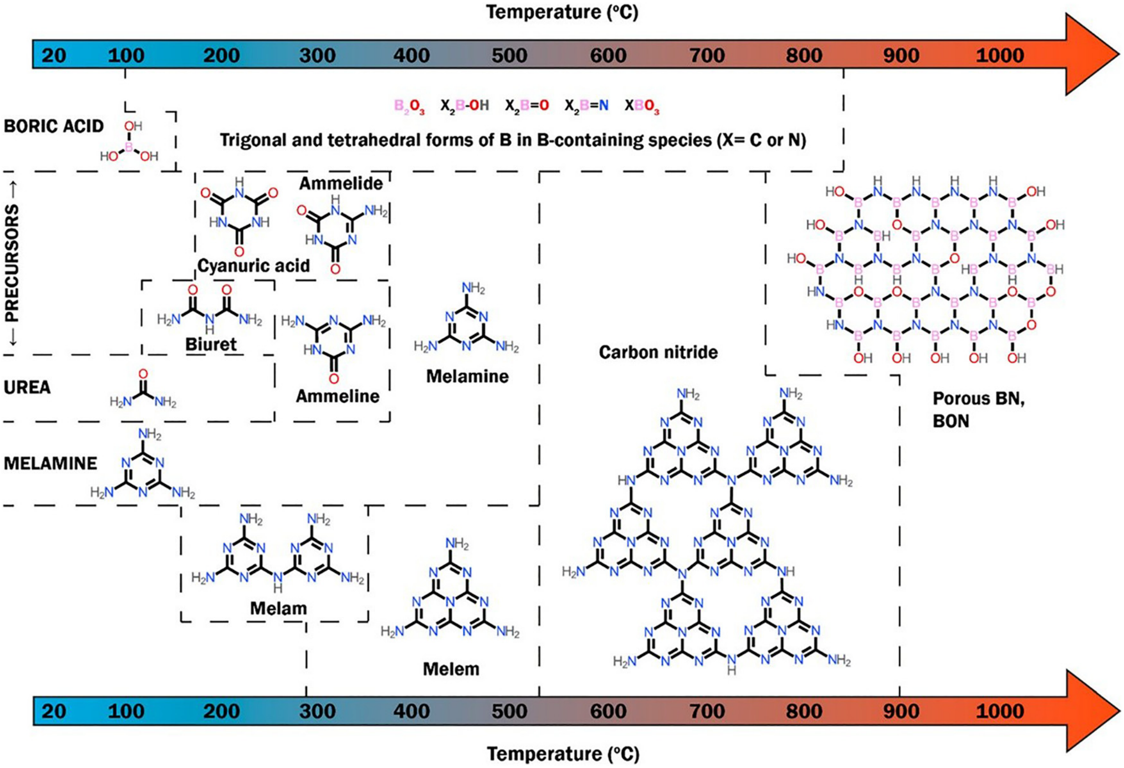

Fig. 5 Proposed species evolution during the formation of porous BN from boric acid, melamine, and urea under N_2 atmosphere up to 1050 °C. Reprinted with permission from ref. 88. Copyright 2021 American Chemical Society.

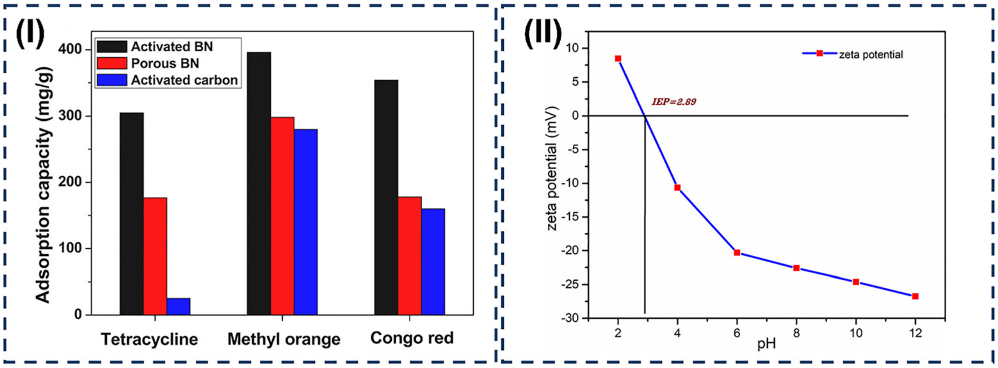

Fig. 6 (I) Comparison of adsorption capacities of the activated BN, porous BN and activated carbon for the tetracycline, methyl orange and Congo red, respectively. Reprinted with permission from ref. 45. Copyright 2013, the authors. Published by Springer. (II) Zeta potential value under different pH. Reprinted from ref. 23, Copyright 2019, with permission from Elsevier B. V.

in the pH, which leads to protonation or deprotonation of functional groups in the molecules. Thus, especially at basic pH values, the adsorption performance of BN for most antibiotics diminishes due to electrostatic repulsion between the negatively charged surface and the deprotonated pollutant.[23,104,105,107] However, as the adsorption primarily occurs in the form of face-to-face stacking through π–π stacking of aromatic groups on the BN surface, high adsorptive loadings can be achieved even if the adsorption is hindered by repulsive interactions.[106,108] Similar effects were observed in the adsorption of dyes with PBN.[106,107,109]

Studies by Liu *et al.*[110] investigated the adsorption of various metal cations on PBN with a specific surface area of 474 $m^2\ g^{-1}$. Very high maximal adsorptive loadings of 120.5 $mg\ g^{-1}$ for Cr^{3+}, 215.8 $mg\ g^{-1}$ for Zn^{2+}, 316.3 $mg\ g^{-1}$ for Pb^{2+}, and 427.1 $mg\ g^{-1}$ for Cu^{2+} were published. Similar to the adsorption of organic molecules, studies on the adsorption performance of the PBNs show a superior adsorption compared to other materials under the same conditions.[45] The ionic nature of the bond type in BN greatly increases its adsorption potential for metal ions.[111] Due to the "lopsided" density characteristics of BN, the material can transfer electrons much more effectively towards metal ions than other adsorbents with non-polar covalent bond types.[45,112] Han *et al.*[113] investigated the competitive adsorption of Cu^{2+}, Cd^{2+}, and Ni^{2+} metal ions in a ternary system. Their work revealed that the distribution coefficient for the adsorption on PBN was $Cu^{2+} > Cd^{2+} > Ni^{2+}$. The arrangement of the metal ions in such an order did not align with any suggestions based on previous studies. Thus, the group proposed that metal ion adsorption of PBN involves multiple simultaneous adsorption mechanisms and does not solely rely on ion-exchange adsorption,[114] adsorption by electrostatic attraction,[115] or the hydrolysis constant of the molecules.[116] In addition, synergistic and antagonistic effects of the metal ions themselves were identified, where ions with higher affinity substituted those with lower ones.[117] XPS studies by Oh *et al.*[118] showed that in binary adsorption experiments, Ni^{2+} and Cu^{2+} metal ions aim to occupy the same B–O active sites on the surface of BN. Especially, B–N and B–O bonds act as attractive adsorption sites for metal ions due to their "lopsided" nature and high electron density.[119] A schematic overview of the adsorption of metal ions on the different adsorption sites of oxygen-doped bundlelike porous boron nitride (OBPBN) is given in Fig. 7. In general, adsorption at the external surface, defect sites, functional groups, and the BN structure are possible.[110] The positively charged metal ions can form strong electrostatic interactions with the oxygen and amino groups at the BN surface.[118,120] Thereby, hydrogen exchange of the metal ions with hydroxyl groups and complexation of the metal ions with surface amino groups can occur.[113,119] Chen *et al.*[97] showed that during the adsorption of Cr^{3+} on PBN also the pH value played a crucial role on the adsorptive loading of the adsorbent due to pH dependent variations in the metal ions oxidation state between Cr^{3+} and Cr^{6+}.

In order to further increase the adsorption performance of PBNs, various modification methods to enhance the surface area, add functional groups, or graft metal species on the surface of the material have been proposed in the literature.[95,121–124] Song *et al.*[122] investigated the effect of amine functionalization with *N*,*N*′-methylene bisacrylamide on the adsorption performance of PBN for Cr^{3+} metal ions. In their work, they showed that the maximum adsorption capacity for functionalized BN greatly increased from ~5 mg g^{-1} (pristine) to ~80 mg g^{-1} despite halving the specific surface area after functionalization due to pore blocking. The group proposed that the performance increase occurred due to the stronger electrostatic interactions of the metal ions with the amine-loaded surface of the functionalized materials. Wang *et al.*[123] studied the effect of metal modification on the adsorption behavior of different antibiotics: ofloxacin (OFL), norfloxacin (NOR), and enrofloxacin (ENR) with PBN nanofibers. As depicted in Fig. 8(a) the authors achieved the best performance with copper-modified BN nanofibers (BNNF) on the adsorption of ofloxacin. Follow-up investigations (Fig. 8(d)) revealed that the copper modification greatly increased the maximum adsorption capacity for all antibiotics, due to improved electrostatic interactions, chelation, and cation bridging interaction between the antibiotics and the adsorbent. Similar studies of Song *et al.*[124] on the adsorption of tetracycline with metal-modified PBN confirm the positive effect of metal loading on the adsorption performance. In their work, Ni modified PBN showed the highest adsorptive loading of 429.6 mg g^{-1}.

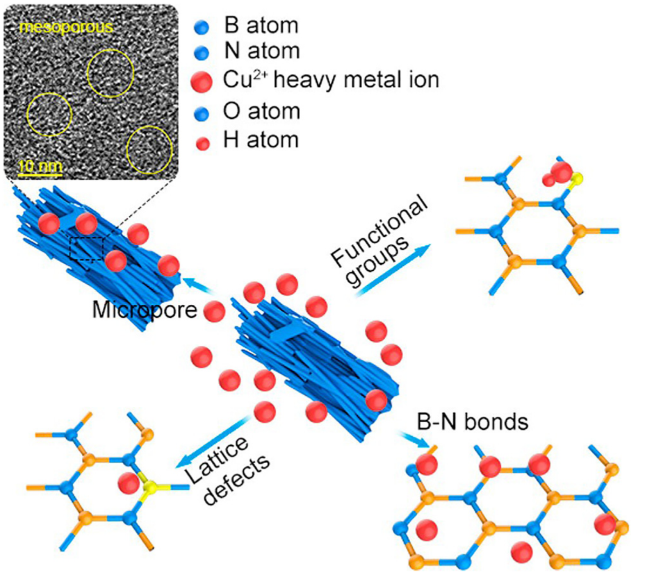

Fig. 7 Schematic illustration of the heavy metal ion removal mechanism with OBPBN. Reprinted with permission from ref. 110. Copyright 2018 American Chemical Society.

In addition to dyes, antibiotics, and metal ions, different BN materials have also been applied in adsorption studies of other pollutants in water, such as phosphate,[125,126] nitrate,[126] fluoride,[127] microorganisms,[128] and PFAS.[129] A comprehensive overview of the different studies carried out on the pollutant adsorption of PBN is given in Table 1.

Apart from its use in pollutant adsorption, BN-based materials and heterojunctions show great potential in the photocatalytic degradation of dyes,[130–139] antibiotics,[130,131,138,140–144] and PFAS.[138,145–148] Hexagonal BN is known as an insulator due to its broad indirect band gap of ~6 eV, depending on the nanostructure.[24] However, due to its inertness, ease of handling, extremely high stability, large surface area, and non-toxicity, it is increasingly applied in photocatalytic heterojunctions acting as a charge carrier acceptor to promote charge separation.[149] Furthermore, the band gap of BN can be easily narrowed up to 1.5 eV by changing the applied synthesis or dope the material.[87,150]

4.2. Cyclic adsorption and regeneration tests

Due to its outstanding properties in pollutant adsorption and photocatalytic degradation, research into the application of PBN in wastewater treatment has been growing rapidly in recent years.[21] A large number of studies show that BN depicts great regeneration and cycle stability even after multiple uses.[22] Depending on the pollutant, there are different regeneration methods for BN. The regeneration of PBN after the adsorption of antibiotics or dyes is primarily based on the high oxidation stability of up to 800–900 °C of the material.[58,182] Here, regeneration occurs by calcination of the pollutant-loaded PBNs at temperatures between 350–600 °C for up to 15 cycles.[95,101,108,123,136,156,157,159,163,168–171] The removal of adsorbed metal ions is usually done by stirring the loaded PBNs in highly acidic[96,110,119,173,180] or basic[121,122] conditions, depending on the investigated metal ion. During regeneration, the surface charge of BN or the charge of the metal complex is changed,

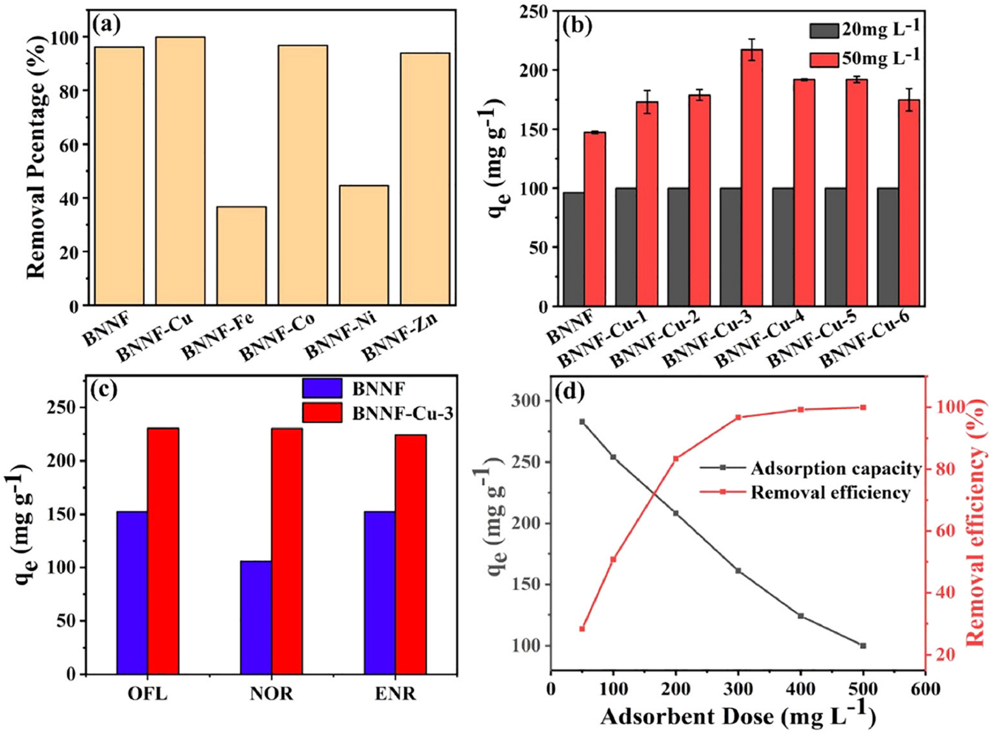

Fig. 8 (a) Removal percentages of various metal ions modified BNNF for OFL, the adsorption capacity of (b) OFL on BNNF-Cu-*X* and (c) OFL, NOR and ENR on BNNF and BNNF-Cu-3, (d) effect of adsorbent (BNNF-Cu-3) dose on OFL removal efficiency and adsorption capacity. Reprinted from ref. 123, Copyright 2022, with permission from Elsevier B. V.

lowering the electrostatic attraction of the ions and the BN surface or leading to repulsion.[23,113] As given in Fig. 9, Wang *et al.*[103] showed that the adsorption capacity for previously synthesized hierarchical porous boron nitride nanosheets (hp-BNNS) is stable throughout 5 cycles in the adsorption of methylene blue (MB) and Rhodamine B (RhB), tetracycline (TC) and Cu^{2+} and Ni^{2+} metal ions. In the dye and antibiotic adsorption (Fig. 9(a)) the group noted an initial decline to 70% adsorption capacity after the first cycle which they attributed to structural changes during regeneration.

Despite showing very high cycle stabilities of PBNs in water treatment, detailed studies on possible changes in the properties of the material during the exposure are just sporadically pursued. However, the few existing studies raise questions regarding the true cycle stability of PBNs.

Marchesini *et al.*[161] investigated the change in structure and chemical properties of different PBNs after dye adsorption for two cycles. In their work, the group synthesized three low temperature (1050 °C) and three high temperature (1500 °C) PBN materials using a template-free approach. As precursors, melamine, urea, and boric acid in the molar ratios 0 : 5 : 1 (BN-U5), 0.25 : 5 : 1 (BNMU0.25 : 5), and 1 : 5 : 1 (BNMU1 : 5) were used. The adsorption tests were carried out by stirring 100 mg BN in 250 mL of a 40 mg L^{-1} RhB solution for up to 4 hours. The first adsorption cycle revealed high adsorption capacities for all materials with a complete removal of RhB within 4 hours. The high temperature samples displayed a faster adsorption of RhB despite their lower surface area. The authors attributed this effect to the higher hydrophobicity and a larger quantity of π electrons of the higher temperature materials, allowing for greater affinity to the adsorbate and a higher stability towards water, which was in agreement with preceding water vapor sorption experiments. After adsorption, the materials were regenerated by calcination in air at 600 °C and analyzed. Nitrogen sorption and XPS revealed varying degrees of degradation and boron oxide formation for all BNs after dye adsorption. As shown in Fig. 10, the pore structure of the low temperature samples was nearly fully lost, whilst the high temperature BNs showed higher stabilities. Nonetheless, for the high temperature samples also a significant reduction in pore volume or specific surface area was noted. Interestingly, the following RhB adsorption tests revealed a similar performance of the already degraded low and high temperature materials as in the first cycle. Marchesini *et al.* attributed this observation to too low dye concentration during the adsorption tests to fully saturate the adsorbents.

A similar loss in specific surface area upon regeneration after 10 adsorption cycles was observed by Liu *et al.*[104] In their work, the authors noted a reduction in the specific surface area of the regenerated porous BNNSs from 1427 m^2 g^{-1} to 940 m^2 g^{-1} without changes in the pore size distribution. The group attributed the change to blocked pores due to residual carbon from the pollutants, which did not completely oxidized. Despite the loss in specific surface area, the materials still showed ∼82% chlortetracycline hydrochloride removal after the tenth cycle.

Other works display similar cycle stabilities for PBN despite noting changes in structure and morphology[125,165,174] or functional

Table 1 Overview of the application of porous boron nitride in water treatment, divided into different sections by pollutants

Pollutant	Adsorbent	C_0 [mg L^{-1}]	t [min]	$q^{\max}$ [mg g^{-1}]	Source
Dyes					
Methylene blue	Porous BNNS	110	180	313	151
	PBN nanocarpet	10	<1	272.4	152
	PBN fiber	100	120	—	153
	PBN whiskers	50–200	60	230	102
	Porous BNNS	10–190	30	413.3	154
	PBN fiber	10	20	631	108
	Porous BCNNS	0–250	60	249	155
	PBN whisker	60	50	13 973	156
	PBN	100	20	471.2	157
	h-BNNS	30	30	17.8	158
	Porous BNNS	100–150	480	750	107
	Fe_3O_4-PBN	50	360	393.5	109
	Porous BCNO	100–150	600	709.5	159
Congo red	3D PBN	110	180	717.5	136
	PBN flakes	10	270	70.9	101
	Porous BNNS	130	180	728	151
	BNNS-OH	50	20	147.5	160
Methyl orange	Porous BNNS	110	180	575	3
	Porous BNNS	10–190	60	—	154
Basic yellow	3D BN	90	180	424.3	136
	Porous BNNS	130	600	556	151
Rhodamine B	PBN flakes	5	30	125	101
	PBN whiskers	3	90	210.1	156
	PBN	40	240	—	161
	BNNS-OH	10–100	120	286.5	160
	PBN	40	10	313.4	157
Antibiotics					
Tetracycline	Porous BNNS	50	600	346.7	23
	Porous BNNS	20–200	180	556	162
	Porous BNNS	50	300	284	104
	PBN fiber	100	1440	229.9	95
	Porous BNNS	200	240	1100	163
	$Ni^{(II)}$-porous BN	20–100	1140	429.6	124
	Porous BN	160	60	322.2	164
	Porous BNNS	200	360	438	105
	Porous BNNS	20	30	263.6	103
	PBN meteorite	20	240	444.4	165
	PBN particle	10–50	4320	390.7	99
	PBN aerogel	5–360	180	1620.4	166
	PBN ribbon	5–300	180	302.1	167
	BN nanoparticle	10–100	4320	502.8	168
	BN aerogel	5	180	322.5	169
	PBN fiber	5–360	180	870.3	170
	Cu–Zn-PBN	50–140	1440	623	171
Oxytetracycline	Porous BNNS	50–60	120	128	107
Ofloxacin	Porous BNNS	30	600	72.5	23
	PBN meteorite	20	240	331.3	165
Cephalexin	Porous BNNS	20	600	225	23
Linezolid	PBN particle	10–50	4320	182.3	99
	BN nanoparticle	10–100	4320	340.6	168
Ciprofloxacin	Porous BNNS	10	120	206	104
Norofloxacin	Porous BNNS	10	120	174	104
Ceftriaxone Na	Ni/Co-PBN fiber	40–120	720	410.9	100
Gatifloxacin	PBN	20–80	15	74.4	98
Fluoroquinolone	Cu-PBN fiber	100	1440	305.3	123
Metal ions					
As^{3+}	Fe_3O_4-BNNS	25	480	30.3	121
As^{5+}	Fe_3O_4-BNNS	400	600	26.3	172
Cd^{2+}	PBN nanoribbon	600	40	649	173
	BN	10	120	15.2	113
Ce^{3+}	Activated PBN	52	360	282	45
Co^{2+}	Activated PBN	52	360	215	45
Cr^{3+}	Activated PBN	52	360	352	45
	O-doped PBN	600	700	119.2	110
	Porous BCN	0–800	1000	1212	97
	h-BN	10	1400	177	120
Cr^{6+}	PBN fiber	20–30	1080	123.3	174
	Amide-PBN	12–20	600	83.8	122
	Modified PBN	100	600	133.4	126

Table 1 (*continued*)

Pollutant	Adsorbent	C_0 [mg L^{-1}]	t [min]	$q^{\max}$ [mg g^{-1}]	Source
Cu^{2+}	O-doped PBN	600	700	426.7	110
	PBN	30–700	120	200	118
	Porous BNNS	50	60	417	103
	PBN microrod	200	5760	365.4	175
	Urichin PBN	50	360	92.9	176
	PBN nanoribbon	500	50	341	173
	BN	10	120	19.7	113
Hg^{2+}	BCNNS	307.8	180	625	119
Ni^{2+}	PBN	30–700	120	95	118
	Activated PBN	52	360	235	45
	Porous BNNS	30	60	280	103
	BN	10	120	9.5	113
	PBN	40–80	480	237.6	177
Pb^{2+}	O-doped PBN	600	700	315.9	110
	Activated PBN	52	360	225	45
	Urichin PBN	50	360	115.1	176
	BCNNS	108	180	211	119
	Porous BNNS	1000	30	845	178
	Porous BNNS	40	180	423.2	96
UO$_2^{2+}$	PBN	250	600	413	179
	DAPy-PBN	0–2250	240	115.7	180
Nd^{3+}	BNNS	50	150	207.3	181
Zn^{2+}	O-doped PBN	600	700	214.3	110
Other pollutants					
Phosphate	Ce-PBN	10–40	1440	24.5	125
	Modified PBN	100	600	106.1	126
Fluoride	Al-porous BNNS	10–100	720	50.5	127
Nitrate	Modified PBN	100	600	68.0	126
Perfluorooctane-sulfonate	PBN	0.4–50	360	0.004	129
Perfluoro-decanoate	PBN	0.4–50	360	0.016	129

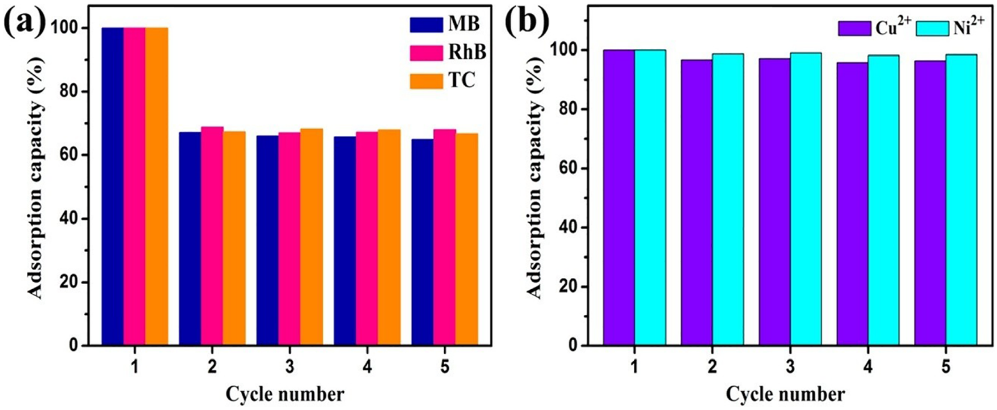

Fig. 9 The recycle ability of hp-BNNSs for (a) MB, RhB, TC and (b) Cu^{2+}, Ni^{2+}. Reprinted from ref. 103, Copyright 2020, with permission from Elsevier B. V.

groups[103,125] for the materials. These results indicate that attributing the water treatment stability and recyclability of PBN to cyclic tests results in a flawed interpretation of the materials actual stability. Without a detailed analysis of the materials porosity or chemical composition cyclic testing seem to indicate high reusability due to effects like insufficient pollutant saturation. Thus, the actual reusability of PBN materials could greatly differ from values indicated in literature. In contrast, a few works[106,154] suggest no changes in the chemical properties of the PBNs after cyclic adsorption experiments and regeneration.

Photocatalytic studies also depict higher stabilities for the used BN species in the heterojunctions.[138,144] However, this can be attributed to the generally higher initial crystallinity and lower porosity of the used BNs in these publications, as most heterojunctions are based on h-BN, which is known for its high chemical stability.[130–132,139] In the photocatalytic degradation experiments, the regeneration of the BN heterojunctions has been achieved by multiple different methods, like pH-induced desorption,[143] magnetic separation,[142] or precipitation and centrifugation methods.[131] Shukla *et al.*[143] used 0.1 M NaOH to regenerate a BN/MoS$_2$/ZnO photocatalyst after CIP adsorption for 5 cycles. The pH-induced desorption was achieved by forming a sodium salt from CIP that desorbed from the catalyst surface. Magnetic separation was applied by Dhiman *et al.*[142] to remove

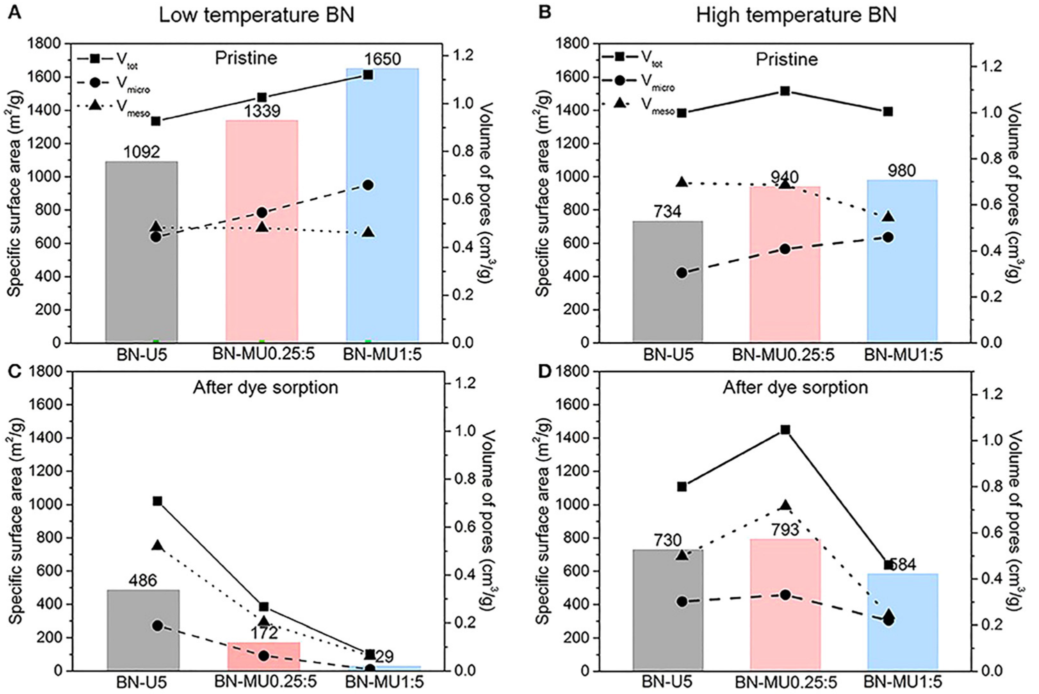

Fig. 10 Changes in the porosity of the BN samples upon adsorption and regeneration at 600 °C in air. Textural parameters derived from N_2 sorption isotherms at −196 °C for porous BN samples: (A) synthesized at low temperature, (B) synthesized at high temperature, (C) synthesized at low temperature after dye sorption and regeneration, (D) synthesized at high temperature after dye sorption and regeneration. Reprinted with permission from 161. Copyright 2019 Marchesini, Wang and Petit.

their BN/Co_2P/$Bi_2O_2CO_3$/magnetic biochar photocatalyst from the TC solution of degradation experiments. Any remaining pollutant was removed by washing the catalyst in an ethanol–water mixture. The group observed cycle stability for up to 6 cycles. Sankeetha *et al.*[131] investigated MB, metronidazole, and cephalexin degradation using a $FeVO_4$/h-BN photocatalyst for 5 cycles. Between cycles the catalyst was removed by centrifugation and washed before reuse.

4.3. The effect of crystallinity on the water stability of boron nitride

Despite the high stability of h-BN towards hydrolysis,[183,184] research on the hydrolytic stability of PBN greatly calls into question its application in wastewater treatment. Multiple publications demonstrate that without special treatment, PBN is not suited to be applied in aqueous media, as a rapid reaction between BN and water takes place.[35,36,153,161]

One of the first research articles demonstrating the low hydrolysis stability of boron nitride dates back to the late 1960s. At that time, Rand and Roberts[185] investigated the synthesis of thin films of BN by chemical vapor deposition (CVD). In their publication, the authors address the already known weathering phenomena of thin film and bulk BN in the presence of water vapor. They observed that storing the prepared thin films in air for more than a week leads to hydrolysis of BN to boric acid and a reduction in stability. A storage in an inert atmosphere, on the other hand, led to no changes.

Almost 30 years later, Matsuda[186] readdressed the problem. During the stabilization and powdering process of previously synthesized BN plates, Matsuda observed a reaction of some prepared BN samples with moisture in the air. By using infrared spectroscopy (IR), the author was able to associate the new signals with the formation of ammonium borate hydrate. Further studies at different reaction temperatures showed that lowering the temperature from 2000 to 1200 °C in 200 K steps steadily decreased the stability of the synthesized BN plates towards moisture.

Similar results were observed by Alkoy *et al.*[57] who compared the crystallinity of t-BN directly after the synthesis and after one year of storage under atmospheric conditions. As displayed in Fig. 11(I), the authors observed a significant change in the materials crystal structure, which was attributed to a reaction with water vapor resulting in the decomposition of t-BN to an ammonium borane compound.

Cofer and Economy[58] investigated the correlation between the d_{002} interlayer spacing of BN and its hydrolytic stability in water vapor-saturated air at 700 °C. They compared 5 different BN samples with interlayer spacings between 0.333 nm to 0.367 nm. The isothermal weight losses of the samples are shown in Fig. 11(II). These indicate a clear correlation between the crystallinity of the prepared BNs and their moisture stability. Hexagonal BN with the smallest interlayer spacing of 0.333 nm depicts the highest hydrolytic stability. With increasing interlayer spacing, however, stability significantly decreases. The authors attributed this trend to smaller grain sizes and weaker atomic bonding due to less densely packed basal planes of the less crystalline BNs. To exclude any effect of oxygen on the stability of the different BNs, Cofer and Economy repeated their investigations in a water vapor

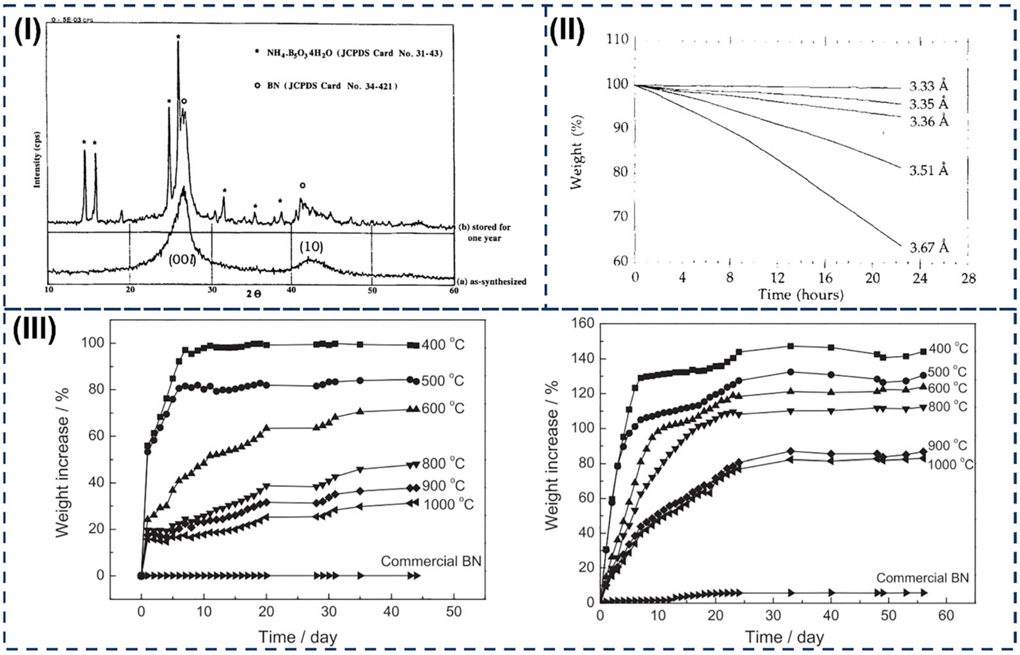

Fig. 11 (I) XRD patterns of the samples (a) as-synthesized, and (b) stored for one year under atmospheric conditions. Reprinted from ref. 57, Copyright 1997, with permission from Elsevier Ltd. (II) Isothermal weight loss in H_2O/air at 700 °C for BN specimens with d(002) from 3.33 Å to 3.67 Å. Reprinted from ref. 58, Copyright 1995, with permission from Elsevier Ltd. (III) Mass growth of BN powders derived from borazine by pyrolysis for 1 h at various temperatures, then exposed to humid air: (a) 25 °C, 65% RH; (b) 40 °C, 90% RH. Reprinted from ref. 187, Copyright 2012, with permission from The Chinese Society for Metals. Published by Elsevier Ltd.

saturated nitrogen atmosphere. Here, no noticeable changes in their findings were observed.

Following on from Cofer and Economy's findings, Cao *et al.*[187] investigated the stability of a-BN towards humidity at room temperature. The group synthesized various BN powders from polymerized borazine at synthesis temperatures between 400 to 1200 °C and exposed them to 65% and 90% relative humidity (RH) over 45 days. During the exposition, they tracked the mass gain of the BNs as shown in Fig. 11(III). The mass gain reduced with increasing pyrolysis temperature. By using X-ray diffraction (XRD) and IR spectroscopy, the authors concluded a reaction of a-BN with water vapor to ammonium borate hydrate, following the proposed reaction mechanism:

$$2BN + 3H_2O \rightarrow B_2O_3 + 2NH_3 \qquad (1)$$

$$B_2O_3 + 3H_2O \rightarrow 2H_3BO_3 \qquad (2)$$

$$H_3BO_3 + NH_3 \rightarrow (NH_4)_2O\cdot B_2O_3\cdot H_2O \qquad (3)$$

Streletskii *et al.*[188] synthesized PBN materials by mechanical activation of h-BN in an argon atmosphere. By changing the applied energy dose (ball mill power multiplied by time) they were able to adjust the surface area and crystallinity of the synthesized PBNs due to a cleavage of the h-BN layers to nanocrystalline rods, increasing interlayer spacing. Later IR analysis of the mechanically activated samples revealed the presence of newly formed bands at 1500 and ~3200 cm^{-1}. Streletskii *et al.* attributed these bands to the formation of boron oxide during air storage at room temperature (RT).

4.4. Effect of chemical properties and structure on the water stability of porous boron nitride

The publications mentioned so far clearly show a general instability of various BN species towards moisture under different conditions.[57,58,185–189] However, these studies have not yet fully addressed the reasons for the instability of less crystalline BN towards water and how its stability can be improved. Likewise, no investigations have been carried out on the effect of liquid water on BN. These questions have been addressed in later works.[35,36,190,191]

The group of Shankar *et al.*[36] performed a detailed investigation on the effect of water and water vapor on two PBN with different surface areas and crystallinities, labelled as high surface area (HSA) BN and low surface area (LSA) BN. Their water stability was investigated by stirring the materials for different time periods in 100 mL of water or 0.1 M HCl at 30 °C. By using a combination of different analyses, the group was able to observe significant changes in the properties of the two BNs. As shown in Fig. 12, XRD analysis revealed an increase in crystallinity for both materials after water exposure. The authors attributed this observation to a decomposition of a-BN in the presence of water and a dissolution of unreacted boron oxide, resulting in a smaller reflex width of the remaining more stable crystalline BN. Additional nitrogen sorption (Fig. 12) investigations reinforced this theory. For the HSA BN, a rapid decrease in specific surface area from 1666 m^2 g^{-1} to ~20 m^2 g^{-1} after 1 hour of exposure was observed, which indicates a removal of porous amorphous and t-BN. For the LSA BN, on the other hand, an increase in the specific surface area was noted for prolonged exposure times or

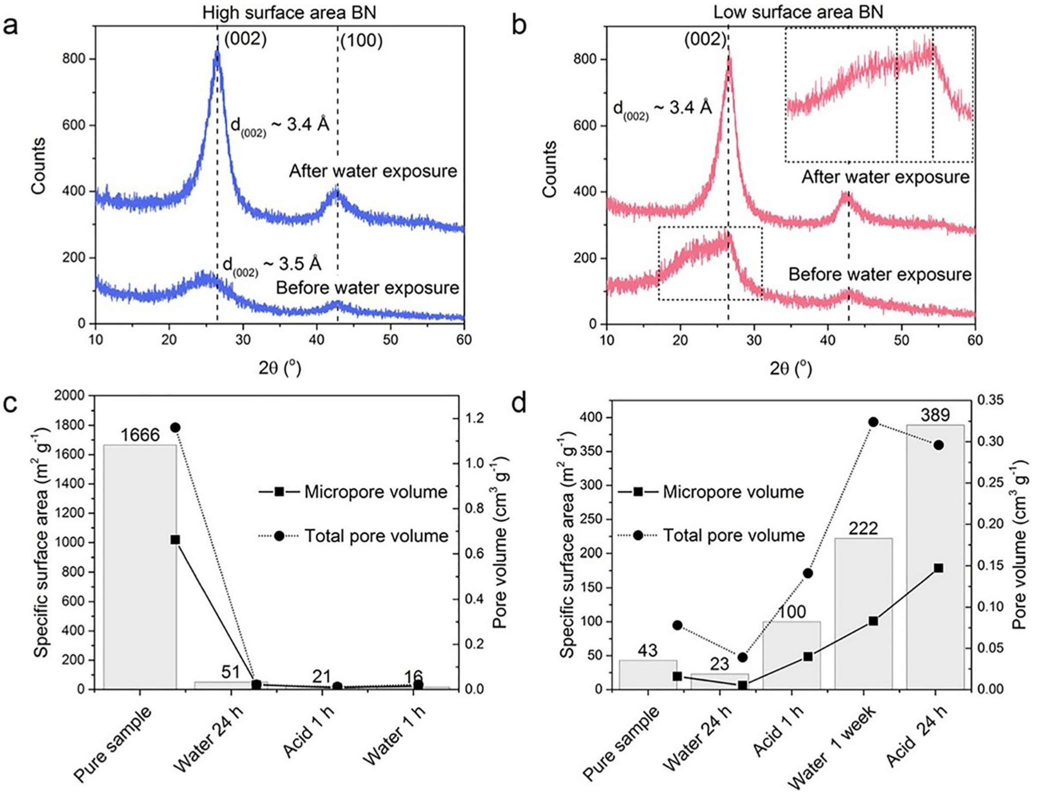

Fig. 12 Comparison of the structure and morphology of the high and low surface area BN samples before and after a 24 h water exposure. (a and b) XRD spectra of the high (a) and low (b) surface area BN samples. (c and d) Bar plots showing the BET equivalent surface areas, total volume of pores, and micropore volume for the high (c) and low (d) surface area BN samples after varying exposure times in aqueous and acidic environments. Reprinted with permission from ref. 36. Copyright 2019 American Chemical Society.

acidic conditions. Shankar *et al.* attributed this result to the aforementioned dissolution of unreacted boron oxide that previously blocked pores in the material. To investigate the effect of water exposure on the chemistry of the BNs, X-ray photoelectron spectroscopy and IR investigations were carried out in the work. These revealed an increase of oxygen in the LSA sample and the broadening of IR bands at ∼3400 and ∼1000 cm^{-1}. The authors attributed these observations to the formation of boron oxide and new hydroxyl group edge functionalization of the BN nanosheets. Shankar *et al.* concluded that achieving water stability for PBN requires a high crystallinity and a low amount of oxygen impurities and defect sites. Following up on their theory, they synthesized a more crystalline PBN material based on the HSA BN at 1500 °C and compared their stability in water vapor sorption. Though the newly synthesized high temperature material displayed a much higher stability, the analysis still showed irreversible chemisorption or reaction with water vapor, as indicated by an open hysteresis loop in the water vapor adsorption.

Florent and Bandosz[190] investigated the effect of water exposure for 2 and 4 days on the chemical and structural properties of boron carbon nitride (BCN). To this end, the group synthesized four different BCN by a template-free synthesis using different molar ratios of boric acid to melamine of 2 : 1 (B_2M), 1 : 1 (BM), 1 : 2 (BM_2), and 1 : 4 (BM_4) at 1000 °C in nitrogen flow (containing 0.5% ammonia). Similar to the results of Shankar *et al.*[36] after the exposure, chemical and structural analysis revealed the complete collapse of the samples porosity, a sharp increase in oxygen impurities due to the formation of boron oxide and hydroxylated species.[190] However, in addition, Florent and Bandosz also identified carbon impurities as reactive sites for hydrolysis. To look deeper into the hydrolysis reaction of the BCNs, Florent and Bandosz investigated the mass loss of the exposed samples by thermogravimetry (TG) analysis coupled with mass spectroscopy (MS). As shown in Fig. 13, the TG analysis revealed three distinct mass losses of $m/z = 16$, 17, or 18 at 80, 180, and 320 °C. By linking these to the hydrolysis products, the authors identified the removal of weak and strongly bonded water as the first two mass losses and the release of ammonia or ammonium as the third mass loss. The mass loss of $m/z = 30$ was attributed to the release of NO.

The research of Bai *et al.*[153] on the water and water vapor stability of porous boron nitride fibers (PBNF) reaffirmed the aforementioned conclusions regarding the stability of BN. After water exposure, the group was able to observe a significant change in the chemical and structural properties of the PBNF. Similar to earlier works, an increase in the oxygen amount due to the formation of boron oxide and hydroxylated species was found. Nitrogen sorption revealed a decrease in the specific surface area of ∼63% pointing towards a structural collapse

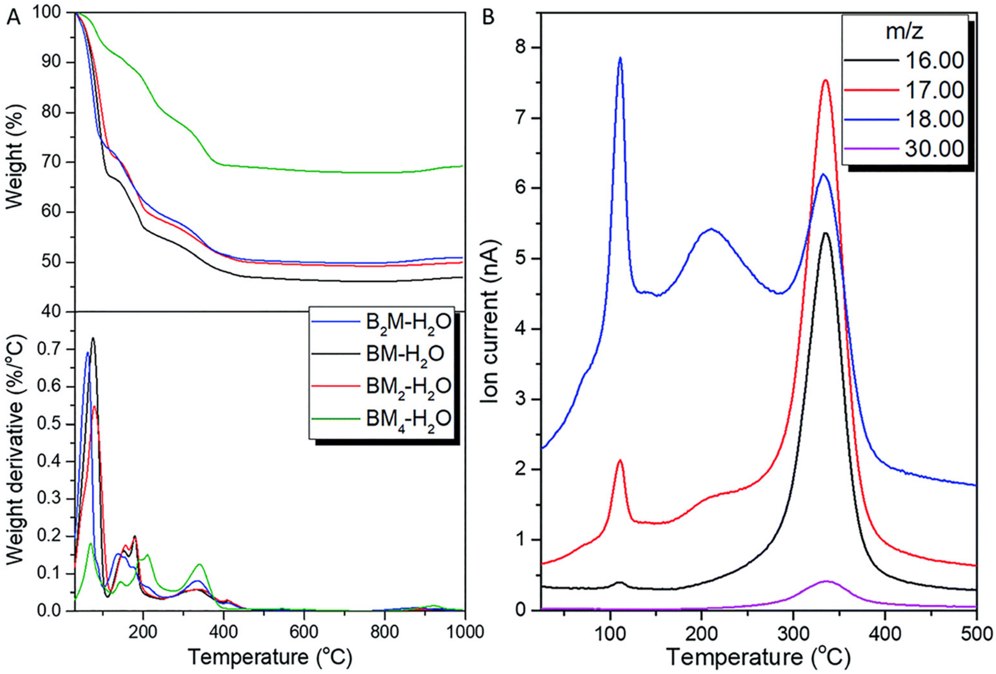

Fig. 13 (A) TG and DTG curves of B_2M, BM, BM_2 and BM_4 after water exposure. (B) *m*/*z* thermal profiles recorded during heating BM–H_2O in helium. Used with permission of Royal Society of Chemistry, from ref. 190; permission conveyed through Copyright Clearance Center, Inc. Copyright The Royal Society of Chemistry 2018.

during exposure. In addition, Bai *et al.* used scanning electron microscopy (SEM) and high-resolution transmission electron microscopy (HRTEM) to investigate changes in the microscopic morphology of the PBNFs due to the exposure. As displayed in Fig. 14, the HRTEM images before water exposure display less defined (002) crystal planes, which indicates a low degree of crystallization. After the exposure, the definition of the (002) crystal planes greatly increases, becoming more regular with enhanced continuity. This reaffirms the theory that water exposure improves the crystallinity of BN due to the decomposition of less crystalline structures. In addition to changes in the PBNF properties, Bai *et al.* also looked into the decomposition mechanism of their material. The group identified the breaking of B–O and B–O–N bonds as the main reasons for the instability, which leads to the formation of reactive B^+, N^+, and NH_4^+ protons and free $O^{\bullet}$ radicals. These further react with the material until an ammonium borate compound is formed, as shown in eqn (4) to (8).

$$O^{\bullet} + H_2O \rightarrow 2OH^{\bullet} \tag{4}$$

$$NH^+ + H_2O \rightarrow NH_4^+ + 3OH^- \tag{5}$$

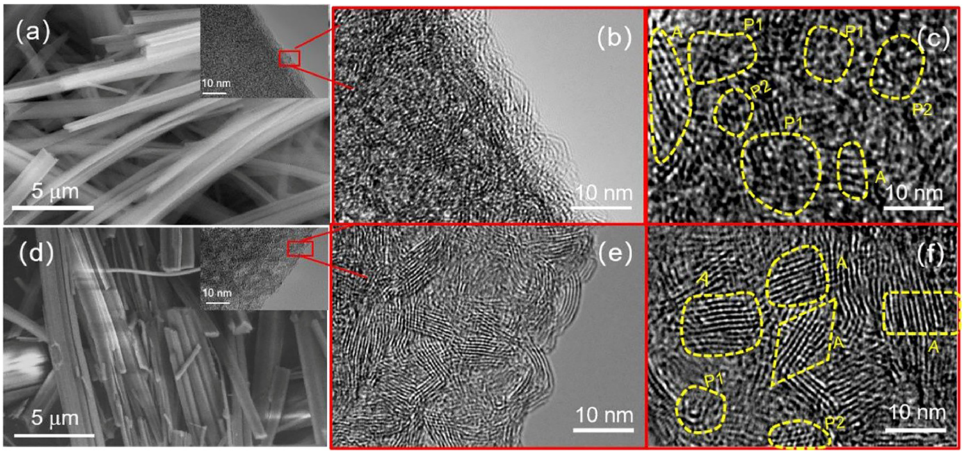

Fig. 14 (a–f) SEM, TEM and HRTEM images of PBNFs before and after water immersion. Dislocation locations and possible P1 and P2 hole types are marked with yellow boxes. The frame shows possible non-parallel and intersecting layer structure models. Reprinted from ref. 153, Copyright 2023, with permission from Elsevier. Published by Elsevier B. V. on behalf of Taiwan Institute of Chemical Engineers.

$$B^+ + H_2O \rightarrow H_3BO_3 + H^+ \tag{6}$$

$$N^+ + H_2O \rightarrow NH_4^+ + OH^- \tag{7}$$

$$H_3BO_3 + NH_4^+ \rightarrow NH_4{\cdot}BO_4^- + H^+ \tag{8}$$

The work of Hojak *et al.*[35] revealed that, in addition to the crystallinity and chemical properties of BN, the pore structure and micropore to mesopore ratio play a crucial role in addressing the stability of PBN. It was shown that in the same sample, mesoporous structures were more stable during water vapor exposure, whilst microporous structures seem to be more stable during liquid water exposure. The authors explained these differences with different reaction mechanisms and limitations occurring in air and water. In air, they proposed a surface reaction of BN with water vapor, leading to the blocking of micropores by the formed products. In water, they suggested that the reaction products are quickly dissolved after their formation, making the material prone to further reactions. Furthermore, they address diffusion limitations in the liquid phase, limiting the rate of reaction in microporous structures and thus a primary decomposition of larger pores in water.

4.5. Strategies to improve the water stability of porous boron nitride

In addition to the already recognized effect of crystallinity,[58,187] other properties such as hydrophobicity,[38] which is influenced by the proportion of oxygen and carbon impurities and defect sites,[36,55,153] and the pore structure[35,161] have been identified to effect the water stability of PBN. To overcome this inherent water instability, new research in optimizing the chemical and structural properties has been carried out in recent years.[93,153,192,193]

Tian *et al.*[192] investigated the impact of shaping of powdered BN to monolithic structures to improve its water stability. By performing a humidity exposure test (>99% humidity) for up to 12 h, the group showed that the stability of the monolithic material could be greatly improved, but no complete stability is achieved as a decrease in the surface area was measured with increasing exposure duration.

Jähnichen *et al.*[93] followed up on the studies of Hojak *et al.*[35] and synthesized purely mesoporous BN. Structural and chemical analysis after water vapor sorption experiments revealed that the mesoporous material displayed no changes in its pore structure, crystallinity, and chemical properties, indicating no reaction with water. Nitrogen sorption analysis (Fig. 15) depicted no changes in the position of the isotherm or in the pore size distribution before and after the sorption experiment for the sample Meso-U3–30 and just slight changes for Meso-U4–30.[93]

L'Hermitte *et al.*[193] looked into the effect of functionalizing PBN with trimethoxy(3,3,3-trifluoropropyl)silane to increase the surface hydrophobicity of the material. Despite successful functionalization and hydrophobization, the water vapor stability of the material was just slightly improved.

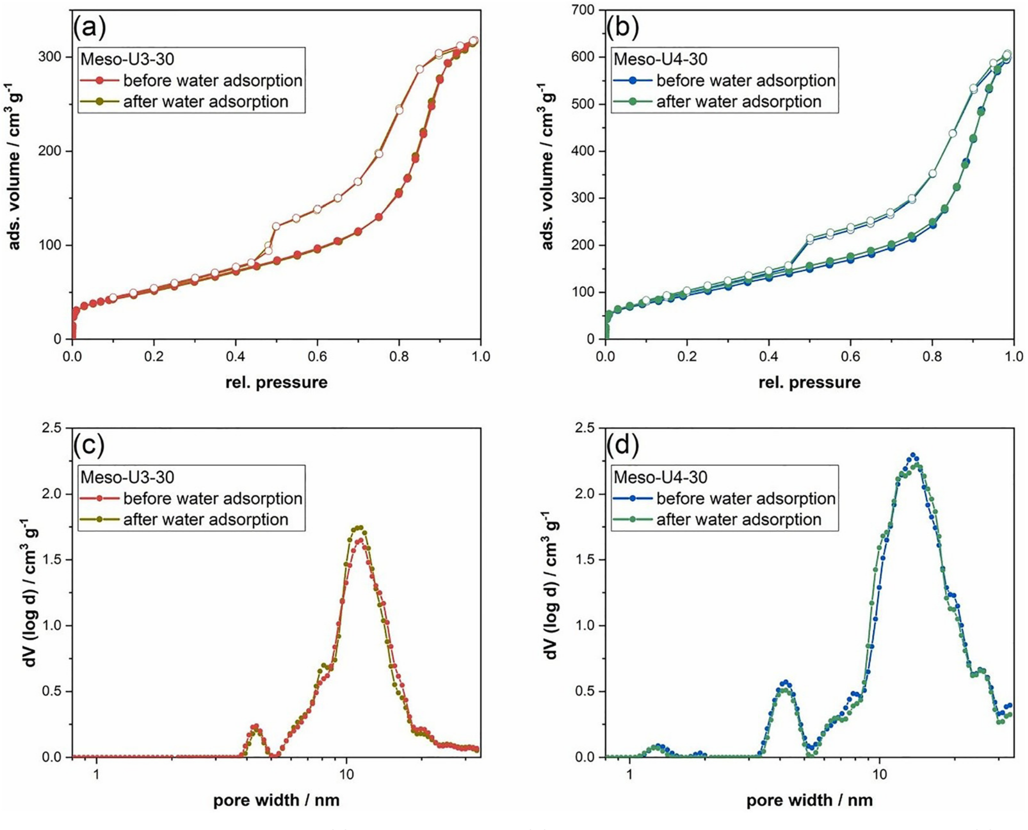

Fig. 15 Nitrogen sorption isotherms of Meso-U3–30 (a) and Meso-U4–30 (b) and pore width distributions of Meso-U3–30 (c) and Meso-U4–30 (d) before and after water-vapor adsorption. Reprinted with permission from ref. 93. Copyright 2023 the authors. Published by Elsevier Ltd.

Bai *et al.*[153] improved previously synthesized PBNFs by post-treatment with a secondary nitriding step in ammonia. This led to an increase in crystallinity and a reduction in oxygen impurities in the material. The authors were able to show that the new material showed much greater stability towards water with only minor changes after exposure. The group also employed the new material successfully in methylene blue adsorption with a 99.5% removal rate after 2 hours. Follow-up studies on the materials properties after dye adsorption were not carried out. The methods discussed above were able to greatly improve the water stability without sacrificing the whole surface area of the material. Nonetheless, a reduction of the surface area due to the modifications was noted on all accounts. Furthermore, despite greatly improving the hydrolytic stability of the PBNs, none of the works reported complete stability. In the future, it will be necessary to carry out more research on further increasing the hydrolytic stability of PBN while maintaining the high specific surface area of the unmodified material. In addition, cyclic exposure and adsorption tests should be carried out to ensure stability over longer periods.

5. Discussion

Porous BN can be a promising material for future applications in water treatment. Due to its unique structural properties and high variability during synthesis, the material can be significantly modified in its morphology, chemical properties, and porosity, thereby enabling a wide range of applications.[24,25,34] Research in water treatment has shown the great potential of PBN in removing a variety of resistant pollutants, including dyes, heavy metal ions, antibiotics, anions, and PFAS.[21,22] These studies depict strong material–pollutant interactions, superior adsorptive loadings, high cycle stability, and easy regeneration of PBNs. However, research on the water stability of PBN calls into question its true stability during water treatment.[57,187] Previous studies[35,36,190] showed that exposure of less crystalline defect-rich PBN to water leads to its decomposition, severely limiting its use for prolonged exposures or repeated cycles. However, this contradiction and potential factors influencing stability during water exposure have received little attention in research to date. There is currently a lack of clear, in-depth studies explaining why PBN exhibits high adsorptive loadings despite indications of decomposition. In the future, detailed studies of the PBN decomposition and pollutant adsorption mechanisms in the presence of different pollutants in addition to theoretical calculations could help to fill these gaps. The work of Marchesini *et al.*[161] proved that solely relying on cyclic adsorption tests is not a sufficient indicator for the stability of PBN in cyclic adsorption experiments. This greatly calls into question the true use of PBN in water treatment applications, as is currently being investigated. Research focuses on synthesizing materials with high surface area to achieve high adsorptive loadings, leading to defect-rich structures susceptible to water. While these materials achieve an effective removal of the targeted pollutants, the released oxidic boron species and ammonia during the decomposition of PBN create severe health risks for organisms.[194,195] A possibility seldom addressed in the literature. In addition, due to a lack of follow-up studies on the materials properties, a false image of the water stability of PBN is conveyed. In order to address these problems in the future, it will be necessary to carry out detailed studies of the material properties after adsorption tests to identify possible instabilities.

Furthermore, studies on achieving water stable PBNs have to be pursued more strongly in research. While current studies[93,153,193] indicate how to improve the water stability of PBN by defect engineering and functionalization, still insufficient stability and a reduction in surface area are noted on all accounts. It is necessary to first focus on realizing the synthesis of a completely water stable material before further pursuing the application of PBN in the adsorption of different organic pollutants. Studies must shift from achieving a high surface area and adsorptive loading first to achieving a stable material before improving adsorption and porosity. Given that the instabilities of PBN to water arise primarily from defect structures the effect of less defect-rich but more stable PBNs has to be considered regarding future applications. Current studies[35,36,153,190] point out that defect structures and hydrophilic functional oxygen or amino groups greatly enhance the adsorbent–adsorbate interactions in water treatment. They can form strong electrostatic interactions with hydrophilic groups of dyes and antibiotics or with metal ions due to their ‘‘lopsided’’ nature and high electron density.[101–103,119] Studies on the stability of PBN suggest that more stable PBNs are much less hydrophilic, which could lead to greatly diminished adsorption performance.[36] Going forward, the approach to research on PBN in water treatment has to be revised in order to fully utilize the unique properties of the material in future applications.

6. Conclusion and outlook

This paper discusses the potential opportunities and problems associated with the use of PBN in water purification. Due to their easy synthesis, high surface area, and advantageous chemical properties, PBNs show great potential in the adsorption and decomposition of pollutants in wastewater such as dyes, antibiotics, metal ions, and other compounds. Various research articles have shown that PBNs depict very high adsorption capacities and good cycle stability. In contrast to those findings, however, studies on the hydrolytic stability of the PBN material itself indicate a high degree of instability when exposed to water. The crystallinity, defect density, amount of oxygen or carbon contamination, pore structure, and hydrophobicity of the BNs were identified as decisive factors. Exposure to water causes less crystalline areas in PBNs to decompose, forming toxic products such as ammonia and oxidic boron compounds. This results in loss of specific surface area and an increase in crystallinity and hydrophobicity for the exposed BNs. However, the instability of the material does not appear to have any detectable influence on its adsorption performance, greatly calling into question previous reported cycle stabilities that lack detailed investigations of the regenerated BNs. Initial research on modifying PBN showed that different strategies to alter the chemical and structural properties can be applied to increase the hydrolytic stability of the material by sacrificing porosity. Nevertheless, so far, no completely water stable PBN material has been reported in the literature that was applied in adsorption experiments. Based on these results, it is necessary to address the following problems in future research:

I. While some initial investigations have already been carried out, it is still unclear how pollutant adsorption experiments affect the stability of PBN in detail. Here, it is important to perform in-depth investigations. The effect on the structure and chemical properties of PBN must be clearly identified, and the effect of different pollutants on the stability must be discovered. It has to be addressed why such high cyclic performances are measured despite clear indications of losses in the specific surface area of the PBNs.

II. The role of the adsorption sites of PBN has to be addressed. Investigations on the hydrolytic stability of PBN have clearly identified defect sites as one of the main reasons for its instability. However, adsorption studies show especially B–O defect sites as important adsorption sites for metal ions. Thus, it is necessary to investigate their role in more detail in adsorption studies. Do they contribute to the materials instability, or does metal ion adsorption block these sites, hindering material decomposition. In addition, it has to be assessed if the materials hydrolytic stability changes with varying pollutant concentrations or pH values.

III. Future studies on water treatment should include tests on the release of oxidic boron species and ammonia during exposure to guarantee the inertness of the BN material.

IV. To safely use PBN materials in prolonged water treatment applications, it is necessary to improve the hydrolytic stability of the material. Thus, the research on synthesizing water stable PBN should be further pursued to discover synthesis methods that increase the materials stability without sacrificing specific surface area.

V. The effect of multiple component adsorption on the performance but also stability of PBN should be investigated in more detail. Initial research indicates that for PBNs, multiple adsorption mechanisms are occurring simultaneously. Here, it is required to investigate the adsorption of artificial multicomponent wastewater and real water samples.

VI. The application of PBNs in photocatalytic degradation experiments and their stabilities in different heterojunctions need to be examined in more detail. Currently, mainly h-BNs with large band gaps are used, limiting the potential of the material.

Conflicts of interest

The author declares no conflict of interest.

Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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