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1. Introduction

Hydrogen is a promising energy carrier that can be stored in large quantities and used to produce power in fuel cells with no harmful emissions. For mobile and stationary power applications, however, the energy density and storage duration of existing hydrogen storage systems are generally inadequate. Theoretically, solid state chemical hydrides could relieve these constraints due to their high available hydrogen density and relatively benign dehydrogenation behaviour and long shelf life. Ammonia borane (AB) and sodium borohydride (SB) as representatives of high capacity chemical hydrides possess 19.6 wt% (ref. 1) and 10.8 wt% (ref. 2) theoretical hydrogen gravimetric capacity, respectively. Strategies to effectively and efficiently release hydrogen from AB and SB at conditions that support practical applications are therefore desired.

AB and SB can release hydrogen through thermolysis and hydrolysis. Hydrothermolysis is also used if heat, besides water, is an essential driving force during dehydrogenation.³

Effective hydrogen release from ammonia borane and sodium borohydride mixture through homopolar based dehydrocoupling driven by intermolecular interaction and restrained water supply[†]

Pei Pei,^a Mark Cannon,^b Grace Quan^b and Erik Kjeang^b*^a

Non-catalytic dehydrogenation of solid-state ammonia borane (AB) and sodium borohydride (SB) mixtures is achieved in this work by water vapor facilitated hydrothermolysis with a maximum release rate of 560 ml $g^{-1} s^{-1}$ at 53.5 °C and dehydrogenation capacity between 7.8 wt% (measured) and 11.7 wt% (theoretical). A novel dehydrogenation pathway is identified, in which intermolecular interaction causes the formation of intermediates and further enables the prevalence of homopolar dehydrocoupling between –BH moieties in AB and SB. The low-temperature heat used in hydrothermolysis enhances the intermolecular interaction, and the oxygen and hydrogen supplied by water vapor promote the dehydrogenation by forming products. This new hydrogen liberation method can uniquely operate without any catalyst, liquid solvent, or high temperature, and provides a compelling solution for disposable hydrogen source application. The present work also unveils for the first time the unique hydride–hydride homopolar dehydrogenation mechanism, which may unlock the true potential of chemical hydride based hydrogen storage systems.

Generally, certain catalytic additives are also needed to facilitate the dehydrogenation process. Commonly used catalytic additives include transition metals,4 metal salts,5 nanostructures6 and protic/aprotic agents.7 Recent attempts of using different hydrides as catalytic additives were also reported, and several metal hydrides have been revealed to facilitate AB dehydrogenation. Kang and co-workers8 showed that mechanical milling of AB with LiH results in 7 wt% hydrogen release at temperatures around 100 °C through the yield of destabilized LiNH2BH3 material. Dovgaliuk et al.9 showed that the integration of AB with $Al(BH_4)_3$ forms a new coordination complex of $Al(BH_4)_3$ -·NH₃BH₃, in which AB can release two equivalent hydrogen molecules at 70 °C through producing $Al(BH_4)_3 \cdot (BH-NH)_n$ intermediate. Similar efforts were also devoted on the combination of AB and SB, motivated by their high theoretical hydrogen densities and benign dehydrogenation. However, intensive study10 has shown that mechanical milling and thermal treatment of an AB and SB mixture cannot release hydrogen below the decomposition temperature of pristine AB at ~ 104 °C. The hydrolytic approach was also employed to dehydrogenate the mixture of AB and SB, and showed that the kinetic improvement of hydrogen evolution mainly derives from the active Co catalyst reduced *in situ* by the mixture rather than the interaction between AB and SB.11 These results indicate that conventional thermolysis and hydrolysis processes are unable to render facile hydrogen release from AB and SB mixtures.

^aFuel Cell Research Laboratory (FCReL), Simon Fraser University, 250-13450 102 Avenue, Surrey, BC V3T0A3, Canada. E-mail: ekjeang@sfu.ca

^bHydrogen in Motion (H2M), Unit 3B 2350 Beta Avenue, Burnaby, BC V5C 5M8, Canada

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Here, we propose an original method to release hydrogen from AB and SB simultaneously through a novel dehydrogenation pathway which is dictated by intermolecular interaction and homopolar dehydrocoupling. No catalyst is required in this process, and the mixing of AB and SB is achieved simply by blending. The method can be categorized as water vapor facilitated hydrothermolysis, however, the functions of heat and water to dehydrogenation are distinct from conventional hydrolysis and thermolysis. Furthermore, only a trace amount of water vapor is utilized instead of liquid water to create a critical water rarefied condition. The exemption from using catalyst and large quantities of liquid water together with lowtemperature heat endues the potential of high effective capacity and facile operation.

2. Experimental

2.1 Chemicals

Ammonia borane (97% purity), sodium borohydride (>99% purity), deuterium oxide (99.9% purity), and silica gel (>99% purity), 200–425 mesh) were purchased from Sigma-Aldrich. Sulfuric acid (98% purity) was purchased from Alfa Aesar. Nitrogen protection gas with purity of 99.999% was purchased from Praxair. The resistivity of deionized water (DI water) used in this research was >18.1 M Ω cm⁻¹. ND₃BH₃ used in the isotope experiment was prepared following the procedures in literature.¹²

2.2 Dynamic dehydrogenation

Dynamic dehydrogenation was conducted on a custom-made Sievert-type apparatus¹³ from 22 to 90 °C with ramp rate of 0.5 °C min⁻¹. A dry glass vial containing 0.169 g ABSB of 3 : 2 mass ratio mixed by mild agitation was loaded into the sample chamber with a water vapor source pre-loaded beneath it. The sample chamber was sealed and leak tested, and then evacuated to 2×10^{-4} Pa for 10 min using a turbo molecular vacuum pump. Heat was thereafter applied while the pressure and temperature were recorded with acquisition frequency of 1 s⁻¹. The pressure and temperature data were converted to gas volume using the ideal gas law, where the sample chamber vapor concentration expressed in relative humidity (RH) was around 49% at the beginning of the dynamic experiment, which was evaluated by directly measuring water vapor pressure at 25 °C.

2.3 Isothermal dehydrogenation

Experimental apparatus for isothermal dehydrogenation measurement is schematized in Fig. S1 (ESI[†]). The ABSB sample was loaded into a dry beaker placed on the bottom of a dry conical flask reactor also containing a water vapor source, in this case is a small piece of wet cotton, separated from the ABSB sample. A thermocouple was attached to the beaker containing ABSB for temperature measurement, and a capillary tube was inserted to extract gas samples for compositional analysis by universal gas analyzer (UGA) prior to purification. The gas released from the reactor was purified by passing it through a condenser (trapping volatile gas), 0.1 N sulfuric acid solution (removing alkaline gas), and silica gel (removing water vapor). The purified hydrogen gas was finally measured in volume by a burette. Prior to measurements, the system was purged by high purity nitrogen for 10 min to eliminate residual air. The superficial volume reading derived from thermal expansion of gas in the system was corrected by subtracting the calibration test data from isothermal measurements. To start the dehydrogenation experiment, the reactor flask containing ABSB and water vapor source was transferred into an oil bath held at 55 °C while monitoring hydrogen evolution.

2.4 Characterization

The compositions of gases and volatile dehydrogenation products were analyzed by an ULT100 universal gas analyzer (UGA) from Stanford Research Systems with maximum resolution of 25 points per AMU. X-ray diffraction (XRD) spectra were collected on a Bruker D8 Advance (Cu K α) with 0.6 mm slit Bragg–Brentano primary optics to identify phase structures of solid residues. Solid state ¹¹B magic angle spinning (MAS) nuclear magnetic resonance (NMR) was measured on a Bruker AVANCE III 400 system to examine chemical states of solid residues. Raman spectra were collected on a Thermo Scientific DXR2 Raman microscope system using 0.1 mW 532 nm excitation laser and 25 µm slit aperture to probe dehydrogenation pathway and mechanism.

3. Results and discussion

3.1 Dehydrogenation performance and influencing factors

The proposed water vapor facilitated hydrothermolysis of AB and SB mixtures (ABSB) was first demonstrated by dynamic dehydrogenation measurement with results shown in Fig. 1. The release of gas started around 30 °C and progressed with a slow release rate until 50 °C. At this temperature, the gas release rate increased drastically, and an eruption of gas was observed at 53.5 °C with a peak release rate of 562.7 ml g⁻¹ s⁻¹.



Fig. 1 Dynamic gas release from the ammonia borane and sodium borohydride mixture heated from room temperature to 90 °C at 0.5 °C min⁻¹. The quantity of gas released was normalized by the initial mass of the mixture.

Further heating to 90 °C did not significantly improve the gas yield, and the total gas released was 1407 ml g^{-1} .

To understand the dehydrogenation behaviour and the influence of the water vapor concentration, isothermal measurements were conducted at 55 °C. Isothermal experiments were performed with three different deionized water sources to create low, medium, and high RH of 17% (ABSB-LRH), 48% (ABSB-MRH), and 97% (ABSB-HRH) in the reactor flask at 55 °C and 1 atm pressure. The RH values reflect the initial levels of water vapor concentration inside the reactor measured by universal gas analyzer (UGA) in ABSB-free calibration tests. The hydrogen release capacities of the obtained ABSB hydrothermolysis isotherms, shown in Fig. 2, were calculated after purification and thermal expansion correction. The hydrothermolysis of ABSB-LRH released 2.90 wt% of hydrogen after 64 min. Similarly, ABSB-HRH released 4.29 wt% after 134 min. Interestingly however, the hydrothermolysis of ABSB-MRH gave a much larger hydrogen release of 7.78 wt% after 149 min. These experiments showed good reproducibility within ± 0.5 wt%. There were no apparent induction periods for any of the measurements, and immediate gas release was observed upon activation with a predominantly consistent release rate until the full capacity was reached in each case. The measurements show that the RH is an important factor during the hydrothermolysis of ABSB, and an appropriate water vapor concentration is required in order to obtain a high degree of dehydrogenation.

Two control tests were conducted, where either mixing (CT 1) or water vapor (CT 2) was omitted intentionally during hydrothermolysis (Fig. 2). Only marginal gas release was observed from each test (≤ 0.5 wt%). Thus, separated AB and SB failed to produce a comparable amount of hydrogen even under optimal (48%) RH, indicating the critical role of AB–SB interaction. Lack of water vapor also largely prevented hydrogen release from ABSB, showing that water vapor presence is required for the desired reaction process to take place. The gas released from ABSB-MRH was analyzed by UGA before being purified with quantitative results appended (ESI, Fig. S2†). The gas detected



Fig. 2 Isothermal hydrogen release from hydrothermolysis of ammonia borane and sodium borohydride mixtures at 55 °C with three different water vapor concentrations of 17%, 48%, and 97% RH. Control tests of isothermal hydrogen release at 55 °C with either the separation of AB and SB (CT 1) or absence of water vapor (CT 2) are also displayed and compared. The capacity of H₂ release was measured in weight percent after purification.

contained 86% (98.2%, if N_2 excluded) hydrogen and only 0.6% (0.7%, if N_2 excluded) ammonia was identified as volatile byproduct. The rest of the gas comprised of nitrogen as protection gas and a trace amount of water vapor.

3.2 Solid dehydrogenation residues

The solid residues from the three isothermal hydrothermolysis experiments were analyzed by X-ray diffraction (XRD), shown in Fig. 3. The XRD spectrum of the pristine ABSB mixture before dehydrogenation was also measured (ESI, Fig. S3†) for reference. The residue from ABSB-LRH hydrothermolysis comprised of NaB₃O₅·2H₂O (JCPDS no. 20-1081) borate together with unreacted AB and SB. The residue from ABSB-HRH hydrothermolysis contained a pentahydrated borate of Na₂B₄O₇-·5H₂O and similarly unreacted AB and SB. In contrast, the XRD spectrum of ABSB-MRH shows a complex pattern identified as a mixture of sodium borates, ammonium borate, and polymeric borates. No unreacted AB or SB was detected in this case by XRD, indicating a complete dehydrogenation of both AB and SB.

¹¹B magic angle spinning (MAS) nuclear magnetic resonance (NMR) was employed to further examine the chemical states of the residues from dehydrogenation (ESI, Fig. S4[†]). In the ABSB-LRH residue, resonances corresponding to unreacted solid AB and SB together with tricoordinated and tetracoordinated boron in borate environment were identified. The ABSB-HRH spectrum resembles that of ABSB-LRH, showing AB, SB, and borate resonances, but with evidence of higher degree of dehydrogenation, especially for SB. In the case of dehydrogenated ABSB-MRH, the resonance corresponding to SB is undetectable, implying a complete dehydrogenation of SB. AB was almost depleted as well, but a weak signal between -22 and -30 ppm remained. This signal has similar chemical shifts as AB, but with apparent new resonance peaks indicating its new identity of AB mobile phase (AB*),14,15 which features a rapidly rotating B-N bond relaxing the extensive dihydrogen network in AB and facilitating the formation of diammoniate of diborane (DADB).16 The AB* mobile phase found in ABSB-MRH was not observed in the other two experiments where the majority of the AB remained unreacted, implying a potentially important



Fig. 3 X-ray diffraction spectra of solid residues from 55 $^{\circ}$ C dehydrogenation of (a) ABSB-LRH, (b) ABSB-MRH, and (c) ABSB-HRH. A phase identification list is provided in the inset.

intermediate role of AB* for complete ABSB dehydrogenation. The chemical shift region of borates is also complex, supporting the existence of ammonium borate¹⁷ and polymeric borate¹¹ in solid products. These results are consistent with the XRD results, representing dehydrogenated reaction products. The collective information about products and isothermal H₂ release capacities indicates that moderate RH values are the key to deep dehydrogenated. In contrast, low or high RH only allows partial dehydrogenation of SB, which is also facilitated by ABSB interaction though, forming regular SB hydrolysates and leaving AB largely unreacted.

3.3 Dehydrogenation pathway

Further efforts were also made to understand the process and transformation during the ABSB-MRH hydrothermolysis by using in situ Raman spectroscopy measurement with spectra evolution of AB in ABSB shown in Fig. 4a for the 2000–3400 cm^{-1} region and in Fig. S5a (ESI^{\dagger}) for the 560–2000 cm⁻¹ region. Initially, all AB vibrational bands are weakened and broadened after heat and vapor are applied, due to the reduction of crystallinity and interaction among components. With the progression of hydrothermolysis, DADB, which is an important intermediate for AB thermolysis, emerges and increases in significance over time. Concurrently, evidence of borate formation is observed. After 3 hours of hydrothermolysis, all previously detected bands from AB and DADB intermediates have vanished, and only weak peaks from borates remain, indicating that the dehydrogenation process is complete. The impact of ABSB interaction on AB can be more clearly observed when comparing the Raman spectra of AB before being mixed with SB (pristine-AB) and at sequential time points in the early stage of dehydrogenation after ABSB being mixed, heated, and exposed to water vapor. The most significant changes were observed in the B-N vibration bands as shown in Fig. 4b, where the intensities are normalized to the strongest peaks of symmetric B-H in each spectrum to reflect relative intensity changes $(I_{\rm BN}/I_{\rm svBH})$. The corresponding B-H band evolution is shown in Fig. S5b (ESI†) as well. Compared to

pristine AB, the intensity of the B-N band was significantly reduced after the mixing of AB and SB, which can be interpreted as weakening and even breaking of B-N bonds. Heating the mixture further attenuated the B-N band, as higher molecular kinetic energy caused by heating enhanced the interaction. AB* mobile phase was possibly formed during this stage. Upon the introduction of water vapor and the commencement of dehydrogenation, the intensity of the B-N band was revived, indicating the strengthening and recovery of B-N bonds. This occurred concurrently with the emergence of a shoulder peak in the B–H band at 2441 cm⁻¹, as shown in Fig. S5b (ESI†), which is related to the formation of DADB. Similar efforts to observe the dehydrogenation of SB in ABSB were also made, with results shown in Fig. S6 (ESI[†]), exhibiting the in situ Raman spectra before ABSB being mixed and after ABSB being mixed and subjected to water vapor and heat. The mixing of SB with AB did not cause any notable change in the Raman spectrum; neither apparent band shifting nor new vibrational bands were seen. However, after applying heat and water, the intensities of all BH₄ bands were attenuated and the combinational band v_{2+4} was suppressed and eliminated, which could imply the alteration of quantity and chemical environment for BH4⁺ due to the occurrence of dehydrogenation interaction and reaction. With the progression of dehydrogenation, the bands related to BH4⁺ vanished gradually, and concurrently, the B3-O and B4-O bands from borates emerged.

In order to unravel the dehydrogenation pathway for ABSB, two isotope modified ABSB-MRH isothermal dehydrogenation experiments were conducted: one with D_2O (99.9 at% deuterium oxide, Sigma-Aldrich) vapor replacing H_2O vapor (ABSB- D_2O); and another one with ND_3BH_3 compound replacing AB in ABSB together with D_2O vapor (AB(D)SB- D_2O). The volatile products collected after 50–60 min of each experiment were examined by UGA in the high resolution mode, and the sections corresponding to hydrogen and isotopes are shown and compared to the spectrum of regular ABSB-MRH (Fig. 5). The UGA spectrum for ABSB- D_2O (Fig. 5a) shows the strong dominance of regular hydrogen (H_2) compared to hydrogen deuteride (HD) originating



Fig. 4 In situ Raman spectra for dehydrogenation of AB in ABSB: (a) spectrum evolution with time in the $2000-3400 \text{ cm}^{-1}$ Raman shift region, where ST indicates stretching mode; and (b) comparison of B–N bands before ABSB being mixed (pristine AB) and after ABSB being mixed (ABSB-mixed), heated (ABSB-heated), and exposed to water vapor (ABSB-*in situ*-5–35 min) up to 35 min. The intensities in (b) are normalized by the corresponding symmetric B–H peaks with arrows indicating the changes compared to the previous spectrum.



Fig. 5 High resolution UGA spectra of hydrogen (H₂) and hydrogen deuteride (HD) released by hydrothermolysis of (a) ABSB using D₂O vapor, (b) ND₃BH₃ + SB mixture using D₂O vapor, and (c) ABSB using H₂O vapor (as reference case). All three experiments used medium RH (MRH).

in part from D₂O, with an H₂/HD integral area ratio of 25.9. The possible dehydrogenation paths for ABSB-D₂O are illustrated in Fig. S7a (ESI[†]). As schematized, when D₂O vapor is involved in the process, HD will be generated by hydrolysis with either AB or SB; on the other hand, when the dehydrogenation happens through the reaction/interaction between AB and SB or through the self-decomposition of AB, only H₂ is generated. The high integral area ratio thus excludes the possibility of any substantial hydrolysis of AB or SB. Furthermore, the self-decomposition of AB can also be excluded, given the absence of SB hydrolysis. This implies that most of the hydrogen gas released originated from AB and SB reaction/interaction. The product from hydrothermolysis of ABSB-D₂O was examined by Raman spectroscopy, and the results shown in Fig. S8 (ESI⁺) indicate the existence of N-D stretching vibration bands, which suggests that water vapor promotes the formation of ammonium in products by providing hydrogen atoms, besides supplying oxygen to borates. A deeper insight into the dehydrogenation mechanism, especially into the role of homopolar dehydrogenation, comes from the analysis of the UGA results for AB(D)SB-D₂O dehydrogenation (Fig. 5b). The possible dehydrogenation paths for AB(D)SB-D₂O are schematized in Fig. S7b (ESI[†]). Besides any HD released from hydrolysis of AB(D)SB, heteropolar dehydrogenation between protonic D in -ND₃ and hydridic H in -BH₃/BH₄⁺ also produces HD. On the contrary, all H₂ generated must come from homopolar dehydrogenation among -BH₂-/-BH₃/BH₄⁻ moieties. Therefore, the UGA results of the AB(D)SB-D₂O isotope experiment unambiguously distinguish the contributions from heteropolar and homopolar dehydrogenations. Here, the reduced H₂/HD integral area ratio for AB(D)SB-D₂O (6.2) compared to that of ABSB-D₂O (25.9) shows the additional contribution from heteropolar dehydrogenation, where the D derives from -ND₃ moieties in AB, as confirmed by the supplementary isotope experiment of AB(D) SB-H₂O in Fig. S9 (ESI[†]). However, the preponderance of H₂ affirms the major role of homopolar dehydrogenation in the overall hydrothermolysis of ABSB.

The mechanism and pathway of ABSB dehydrogenation by vapor facilitated hydrothermolysis can be collectively inferred

from the above results and are schematically illustrated using AB(D)SB as example to differentiate between homopolar and heteropolar dehydrogenations, as shown in Fig. 6. Upon blending of AB and SB, their interaction weakens and even breaks B-N bonds and relaxes the dihydrogen bonding network in AB. This process is greatly enhanced by mild heating of the mixture (e.g. at 55 °C) and eventually enables the formation of DADB. Evidence also indicated a higher reactivity of SB in ABSB. These observations may be attributed to the alteration of electron structure and distribution in AB and SB through interacbetween $-BH_3/BH_4$ moieties, e.g. electrostatic tions interaction, in the presence of water molecules. DADB is also generally seen as an intermediate during thermolysis of AB at temperatures above 80 °C. However, the present ABSB dehydrogenation process does not follow either of the regular pathways of thermolysis or hydrolysis, as can be expected due to the relatively low temperature and absence of catalyst and liquid water in this case. Instead, a new dehydrogenation pathway is observed at this stage as shown by the isotope and in situ Raman experiments. The new pathway mostly relies on homopolar dehydrogenation, specifically through dehydrocoupling between -BH2-/-BH3/BH4⁻ moieties contained in DADB, AB, and NaBH₄. Homopolar dehydrogenation is counterintuitive given the repulsive force between the negatively charged moieties. However, emerging evidence suggests an important, common role of homopolar interaction during dehydrogenation of chemical hydrides. According to the study by Wolstenholme et al.,¹⁸ even for the regular thermolysis of pure AB homopolar B-H···H-B interaction besides the well-known interaction of N-H···H-B contributes a substantial portion of dehydrogenation when the distance between B-H···H-B has been shortened by melting. In the present case, the quantity of HH from homopolar dehydrogenation is about $\sim 6 \times$ greater than the HD amount from heteropolar dehydrogenation. Furthermore, the derivation of D for heteropolar dehydrogenation is more likely from $-ND_3$ moieties than from water (D_2O) molecules. This is distinct from regular hydrolysis, in which heteropolar dehydrogenation is the dominant process with a significant portion of hydrogen supplied by water. Nevertheless, water is still essential during the dehydrogenation. Besides assisting the formation of DADB, water molecules also provide oxygen to form borates and hydrogen to form ammonium, which enables the formation of products and thus facilitates the process of dehydrogenation. Importantly, the amount of water vapor also determines the approach and extent of dehydrogenation, which is favoured by moderate RH levels. With insufficient RH, the available water vapor is unable to facilitate the homopolar interaction and to sustain the formation of products by providing adequate O and H atoms, whereas excessive water vapor will rapidly deplete SB through regular hydrolysis into sodium borates, and thus deprive the interaction between AB and SB and prevent further dehydrogenation of ABSB.

The regeneration of spent fuel and impurities in products are thorny issues for AB and SB hydrogen storage materials. Developing effective methods to solve these problems is a hot topic, and promising progress has been made.¹⁹ Theoretically, the techniques developed for hydrolytic dehydrogenation



Fig. 6 Proposed pathway for dehydrogenation of AB(D)SB by hydrothermolysis. The methods used to identify the intermediates, products, and processes are indicated in brackets.

should be also applicable for the regeneration and impurity suppression of ABSB. On the other hand, the new homopolar based dehydrogenation mechanism of ABSB renders possibilities of new strategies to simplify spent fuel regeneration and impurity elimination. As aforementioned, the water in conventional hydrolytic dehydrogenation commits to supply H/ O atoms for dehydrocoupling and also for solid product formation. Under the new mechanism, the main role of water is more simple, which is to promote the formation of products. Therefore, it is rational to hypothesize that other gaseous reagents could be utilized to reduce the thermodynamic stability of the products without compromising dehydrogenation. Although the ammonia emission from ABSB dehydrogenation is low (0.7 vol%), further reduction and even elimination of ammonia are feasible through maximizing homopolar dehydrogenation and avoiding side reaction. These could be achieved by optimizing the parameters of hydrothermolysis. The gap between theoretical capacity and measured capacity could also be narrowed by parameter optimization. Further research is needed to systematically investigate the optimal AB to SB ratio and optimal vapor concentration range under a certain AB/SB ratio.

4. Conclusion

The present work revealed for the first time the phenomenon and mechanism of simultaneous and interactional dehydrogenation of ammonia borane and sodium borohydride at near ambient temperature without using any catalyst and liquid solvent. Mixing, moderate humidity, and mild heat are the only requirements for liberation of hydrogen. Major hydrogen release was demonstrated at 53.5 °C with peak evolution rate of 562.7 ml g⁻¹ s⁻¹ via dynamic dehydrogenation experiment. The hydrogen gas was of

high quality with less than 1% of ammonia as impurity. Water vapor concentration was a decisive factor for the approach and degree of dehydrogenation, and the highest extent of dehydrogenation (7.78 wt%) was achieved with moderate relative humidity. In situ Raman, isotope experiments, and thorough analysis of the solid products unraveled the novel homopolar based dehydrogenation mechanism, which is driven by intermolecular interaction with AB* and DADB as intermediates. Distinct from conventional hydrolytic dehydrogenation, the role of water vapor is to promote the formation of products and the interaction rather than participating in dehydrocoupling. The ratio between homopolar and heteropolar dehydrogenation was about 6:1. Based on this, a theoretical maximum capacity of 11.7 wt% was predicted. Overall, this work validated the possibility of dehydrogenating chemical hydride mixtures through intermolecular interaction. The low-temperature heat, minimal water use, and absence of catalyst for hydrogen liberation are key advantages for ABSB as an excellent disposable solid state hydrogen source with simple operation, high energy density, and rapid kinetics. Moreover, the homopolar based dehydrogenation mechanism could render the possibilities of better spent fuel regeneration and impurity elimination strategies. Further study based on this is suggested.

Conflicts of interest

A patent application has been filed based on this work (US 62783620; PCT/CA2019/051841).

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