

Article

In Situ Preparation of 2D Co-B Nanosheets@1D TiO₂ Nanofibers as a Catalyst for Hydrogen Production from Sodium Borohydride

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Abstract: In this study, 2D Co-B nanosheet-decorated 1D TiO₂ nanofibers (2D Co-B NS-decorated 1D TiO₂ NFs) are synthesized via electrospinning and an in situ chemical reduction technique. The as-prepared catalyst showed excellent catalytic performance in H₂ generation from sodium borohydride (SBH). When compared to naked Co-B nanoparticles, the catalytic activity of the 2D Co-B NS-decorated 1D TiO₂ NFs catalyst for the hydrolysis of SBH is significantly enhanced, as demonstrated by the high hydrogen generation rate (HGR) of 6020 mL min^{−1} g^{−1} at 25 °C. The activation energy of hydrolysis was measured to be 30.87 kJ mol^{−1}, which agreed with the reported values. The catalyst also showed good stability. Moreover, the effects of SBH, catalyst concentration, and temperature on the catalytic performance of 2D Co-B NS-decorated 1D TiO₂ NFs were studied to gain a comprehensive understanding of the dehydrogenation mechanism of SBH. Based on these findings, we conclude that 2D Co-B NS-decorated 1D TiO₂ NFs are effective catalytic materials for the dehydrogenation of SBH.

Keywords: hydrogen generation; sodium borohydride; cobalt-boride nanosheets; TiO₂ nanofibers; electrospinning; chemical reduction



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1. Introduction

Hydrogen, due to its wide availability and eco-friendly nature, can serve as a future clean energy carrier [1–3]. Hydrogen has gained tremendous popularity as a green fuel option for fuel cells during the past recent years. Metal hydrides are potential solutions to address the challenges of transport and storage. Sodium borohydride (SBH; NaBH₄), a metal hydride, has attracted a lot of interest from the researchers because it offers several advantages over other hydrogen storage materials, including greater control over H₂ production rate, purity, safe production method, ability to recycle the by-product NaBO₂ into borohydride, and potentially a lower temperature at which hydrogen releases [4–6]. Also, it has superior physicochemical features, such as non-flammability, low molecular weight (37.83 g mol^{−1}), high hydrogen capacity (10.8 wt.%), high room-temperature stability in a solid state, high stability in alkaline aqueous solutions, and secure and reliable hydrogen release via hydrolysis. Although it is counterproductive, an alkaline SBH solution frequently undergoes hydrolysis due to the key role NaOH plays in inhibiting SBH self-decomposition.

In addition to NaOH, catalysts are other options to promote a favorable outcome in this reaction. An effective and sturdy catalyst is required to regulate hydrogen generation during the hydrolysis of SBH. Initially, precious metals emerged as prospective catalysts for generating H_2 from dehydrogenation reaction at room temperature [7,8] because of their stability, longevity, outstanding efficiency, and resistance to deactivation [9]. However, due to their scarce availability and high cost, it has become of the utmost importance to investigate their substitute catalysts that are cheap and have the potentials to accelerate reaction rates while simultaneously reducing threshold activation energies. Thus, researchers are now focusing increasingly on abundant earth metals, such as transition metals like copper, cobalt, and nickel that are capable of catalyzing the dehydrogenation process of SBH [10–12].

Various studies have shown that catalysts constituting cobalt are most effective in hydrolyzing SBH. Among Co-based catalysts, cobalt-boride (Co-B) catalysts show unique features in the hydrolysis process of SBH. In particular, amorphous Co-B catalysts are superior to crystalline Co-B, Co metal powder forms, and other precious catalysts in terms of H_2 generation rate [9]. Co-B is a distinguished candidate due to a number of unique features, including its isotropic structure, a large concentration of coordinative unsaturated sites, and its stable chemical properties.

Recent research has shown that nanoscale Co-B materials with large surface areas and porosities facilitate the transfer and interaction of reactant molecules, leading to increased catalytic efficiencies as compared to conventional Co-B [9]. Naked nano-Co-B is readily susceptible to oxidation in air. It is particularly prone to aggregation and has a high degree of instability. Furthermore, naked nano-Co-B, being ferromagnetic, adheres themselves to the magnetic stirring rod during the hydrolysis process. Nano-Co-B can be stabilized when placed on the support matrix. Nano-TiO₂ has many useful properties as a catalyst and photocatalyst, including excellent chemical resistance, non-toxicity, and efficiency. H_2 production via AB hydrolysis has been reported by Yousef et al. [13], utilizing nano-TiO₂ as a supporting framework for copper (0) as an effective and recyclable catalyst. It has also been shown that palladium immobilized on TiO₂ nanoparticles is extremely effective, can be isolated, and is recyclable in AB hydrolysis, as demonstrated in the work of Rakap et al. [14]. Our research group has seen encouraging results by employing TiO₂ nanofibers (TiO₂ NFs) as support matrixes. NFs possess a high surface area and less aggregates than other nanostructures because of their high axial ratio and less grain boundaries. In addition, the surface area of NFs is almost twice than that of presently available thin film materials [15].

One-dimensional NFs and two-dimensional nanosheets (NS) stand out among the many different morphologies due to evident improvements in particle performances that they offer during catalysis and various other applications [16]. Due to their unique structures with high specific surface areas, they significantly improve the efficiency of the catalytic process. In this paper, we examine the catalytic activity of 2D Co-B NS-supported on 1D TiO₂ NFs synthesized via electrospinning and an in situ chemical reduction technique. The catalytic activity of the produced catalyst is shown to be superior for the release of H_2 from SBH.

2. Results and Discussion

It is well known that organic and inorganic nanofibers produced by electrospinning have their own distinctive nanoporous structures. FESEM images of pure TiO₂ nanofibers and CoAc/TiO₂ produced via the in situ chemical reduction of CoAc with SBH are displayed in Figure 1. The continuous, smooth, and beads-free surfaces of the pristine TiO₂ NFs can be observed in the image shown in Figure 1a. The FESEM in Figure 1b clearly reveals very thin horizontal and vertical Co-B NS attached to the surface of the TiO₂ NFs. The average size of the prepared NFs is 221.60 nm (inset in Figure 1b). The formation of the structure that consists of Co-B NS@TiO₂ NFs can be inferred by visual inspection of the surface of the TiO₂ NFs covered with very thin Co-B NS, as shown in Figure 1c,d. These

structural features are useful to provide a greater number of catalytic active sites on the surface of the Co-B catalyst, allowing better adsorption of reactants. When CoAc and TiO₂ NFs are mixed, the pores in the TiO₂ NFs are filled with CoAc. When SBH is introduced, the CoAc on the nanoporous surface is converted into Co-B NPs. As the reaction continued, the newly generated nanoparticles acted as nucleation sites, and Co-B NPs grew on the surfaces of the TiO₂ NFs. This process continued until the whole CoAc was successfully converted to Co-B. Finally, the 2D Co-B NS grew on the surfaces of 1D TiO₂ NFs.

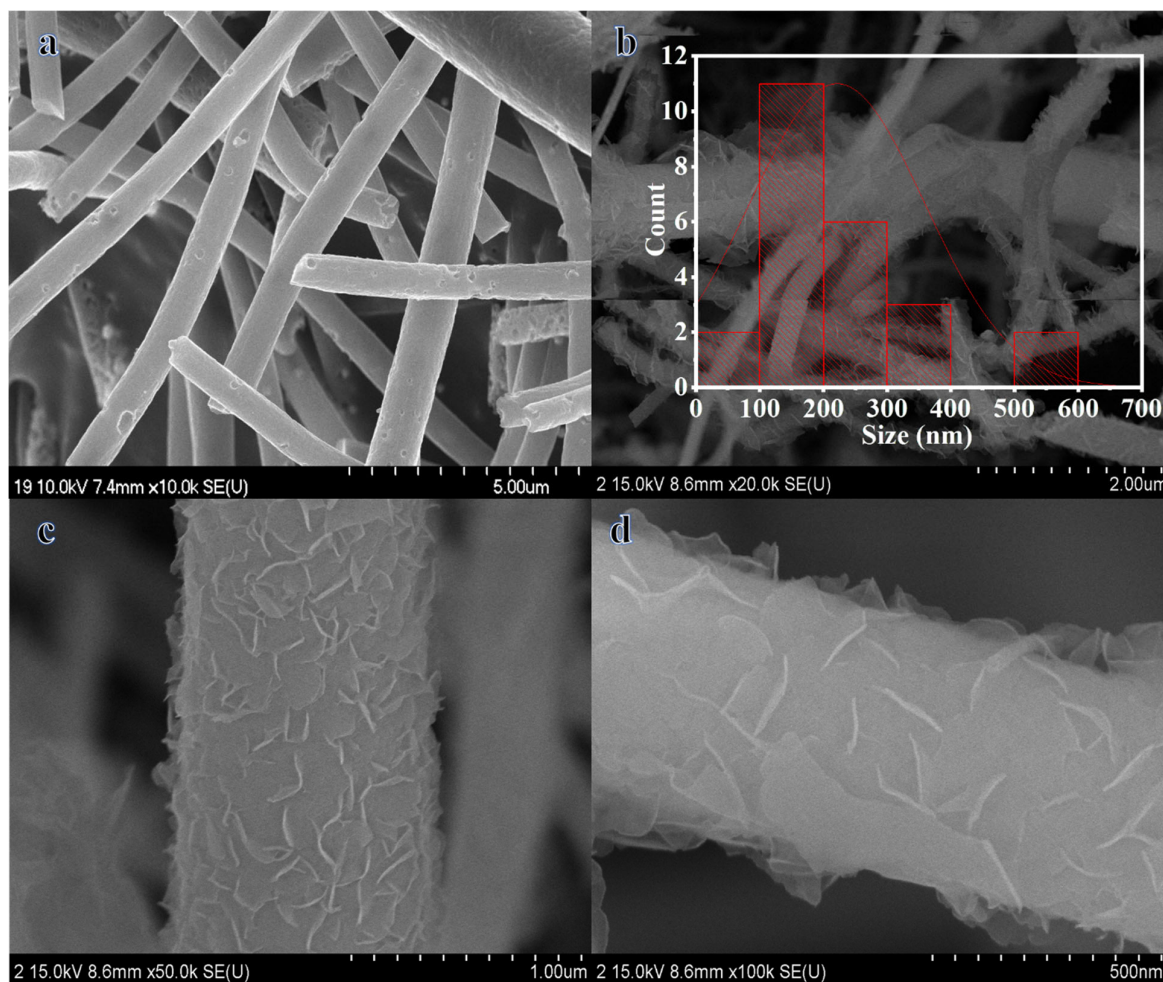


Figure 1. (a) FESEM image of 1D TiO₂ NFs, and (b–d) FESEM images of 2D Co-B NS@1D TiO₂ NFs with different magnifications.

XRD studies were performed to assess the phase structures of pristine 1D TiO₂ NFs and 2D Co-B NS@1D TiO₂ NFs (Figure 2). Pristine 1D TiO₂ NFs exhibited an XRD pattern characteristic of mixed phases of anatase and rutile, as illustrated in Figure 2. It was observed that the peaks at 2θ values of 27.45°, 36.12°, 39.35°, 41.30°, 44.18°, 54.37°, 56.63°, 62.83°, 64.15°, 69.00°, and 69.94° agreed with the (110), (101), (200), (111), (210), (211), (220), (002), (310), (301), and (112) planes of the rutile phase (JCPDS#21-1276). The similar spectra also show peaks at 2θ of 25.28°, 37.88°, and 48.19° for the anatase phase (JCPDS#21-1272) and matches with the (101), (004), and (200) crystal planes. Sharp diffraction peaks in 1D TiO₂ NFs indicate their extremely crystalline natures. It is evident from the diffraction peaks of Figure 2 that the 2D Co-B NS@1D TiO₂ NFs comprise entirely well-crystallized tetragonal TiO₂ (Anatase; JCPDS # 21-1272) and (Rutile; JCPDS # 21-1276). However, no peak is seen for Co-B, which may be due to its amorphous nature.

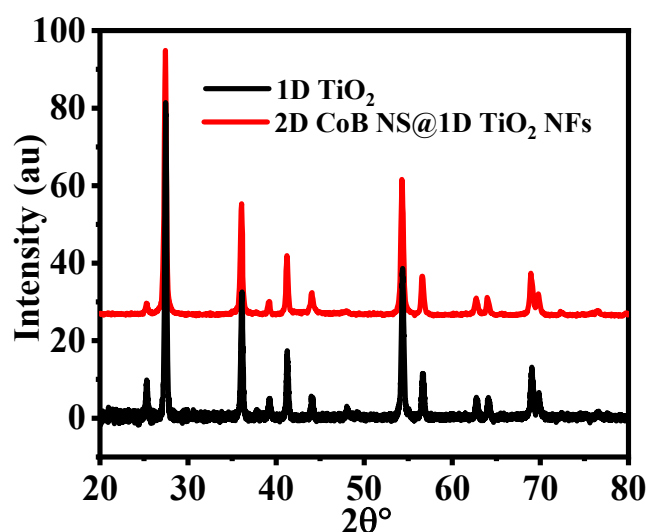


Figure 2. XRD pattern of 1D TiO₂ NFs and 2D Co-B NS@1D TiO₂ NFs.

XPS was employed to confirm the existence of Co and B (Figure 3). The XPS survey spectra of the as-prepared catalyst is depicted in Figure 3a. It can be observed from the figure that the as-prepared catalyst consists of elements Oxygen (Figure 3b), Titanium (Figure 3c), Boron (Figure 3d), and Cobalt (Figure 3e). Figure 3e depicts two peaks corresponding to Co 2p of Co²⁺ states occurring at the binding energies of 782.10 eV and 797.80 eV, respectively [17–19]. Moreover, three satellite peaks of 788.19, 801.33, and 803.94 eV were observed [2,3]. This suggested that the oxidation of the catalyst's surface may have occurred during the sample preparation for analysis. The presence of B was confirmed through the observation of a peak at a binding energy of 191.2 eV (Figure 3d) associated with the B1s level. This peak was shifted by +3.5 eV in comparison to pure B (187.8 eV) [20], suggesting an electron transfer from B to the vacant d-orbital of metallic Co that may have formed linear chains containing B-B bonds [19]. Furthermore, the presence of borate (B³⁺) in the formed boron cannot be excluded [21]. This electron transfer results in the surplus of electrons in Co and a deficiency of electrons in B [22,23]. Furthermore, the estimations of elements and compounds in the produced NFs for Titanium Dioxide (TiO₂), Boron (B), and Cobalt (Co) were found to be 63.69 wt%, 6.37 wt%, and 29.93 wt%, respectively.

The catalytic performance of the synthesized catalyst was evaluated by adding 5 mg of catalyst to a solution containing 26 mM and 0.25 M NaOH at 25 °C. The volume of H₂ that was generated by the dehydrogenation of SBH was continuously monitored. Figure 4a depicts the volume of H₂ achieved by the Co-B NPs and 2D Co-B NS@1D TiO₂ NFs catalyst. As shown in figure, the 2D Co-B NS@1D TiO₂ NFs catalyst generated a higher volume of H₂ as compared to Co-B NPs and pristine 1D TiO₂ NFs catalyst. Lu et al. [24] studied the hydrolysis of the dehydrogenation of SBH using Co-B supported on different substrates (Al₂O₃, TiO₂, and CeO₂) and compared it with naked Co-B. The obtained results indicated that supported Co-B produced a high volume of H₂ as compared to the naked Co-B because Co-B was supported four times higher than the naked Co-B catalyst. Furthermore, Co-B-supported TiO₂ producing 6738 mL min^{−1} g^{−1} of H₂, which demonstrated higher catalytic performance as compared to the Co-B-supported Al₂O₃, which produced 5495 mL min^{−1} g^{−1}, and Co-B-supported CeO₂, which produced 4717 mL min^{−1} g^{−1}. Also, the HGR was found to be 6020 mL min^{−1} g^{−1} for 2D Co-B NS@1D TiO₂ NFs, which was higher than that of the 2672 mL min^{−1} g^{−1} naked Co-B (Figure 4b). This may have been due to the increased number of available active sites on the surfaces of the 2D Co-B NS@1D TiO₂ NFs.

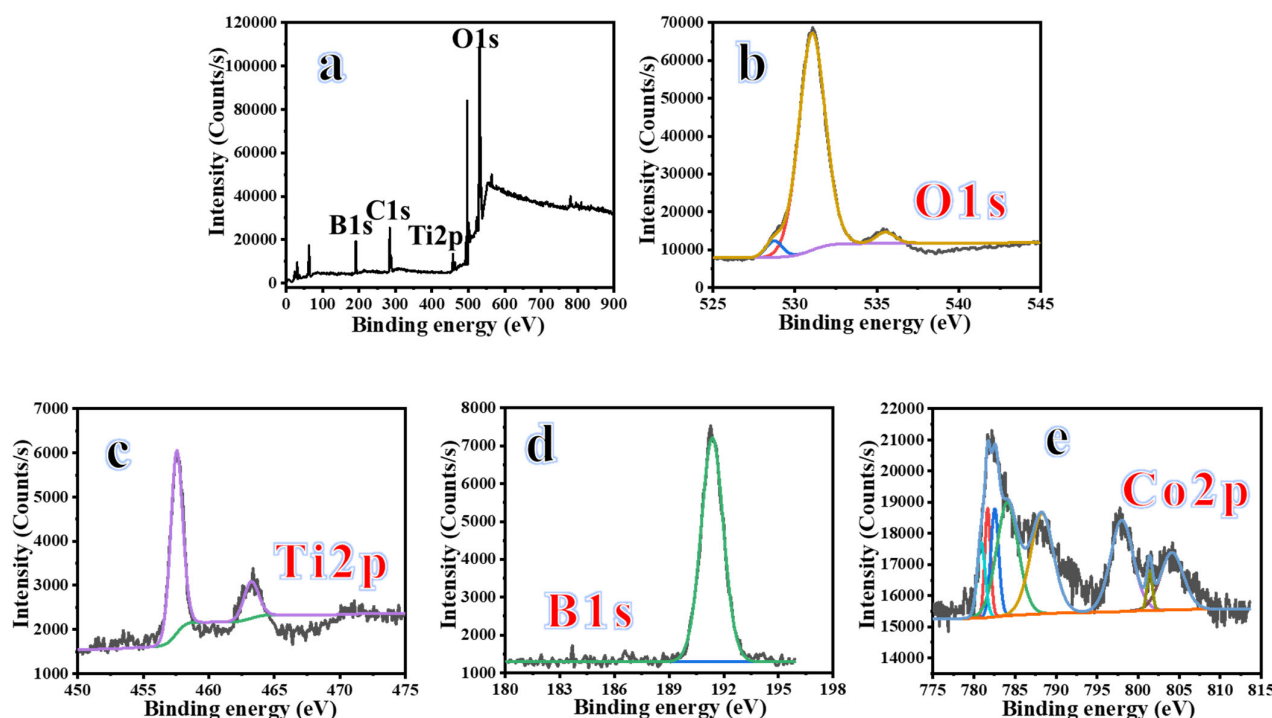


Figure 3. (a) XPS survey of 2D Co-B NS@1D TiO₂ NFs; (b) Oxygen, O; (c) Titanium, Ti; (d) Boron, B; and (e) Cobalt, Co.

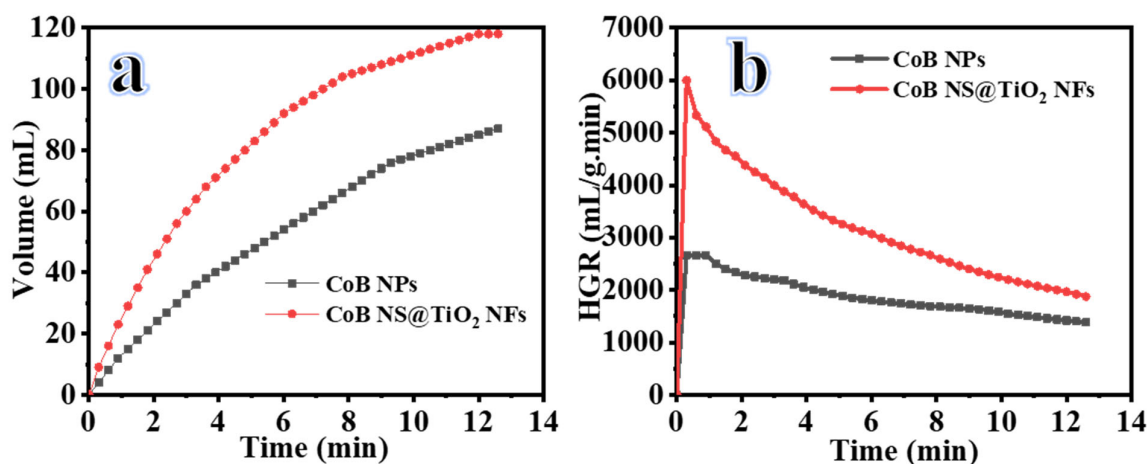


Figure 4. (a) H₂ volume vs. time for dehydrogenation of SBH and (b) HGR using naked Co-B NPs and 2D Co-B NS@1D TiO₂ NFs catalysts.

2.1. Influence of 2D Co-B NS@1D TiO₂ NFs Loading

Figure 5 shows the volume of H₂ production versus time by varying 2D Co-B NS@1D TiO₂ NFs catalyst loading from 5 to 20 mg while maintaining the other parameters (26 mM of SBH, 25 °C, and 1000 rpm) to be constant. The volume of H₂ rises rapidly with increased catalyst loading (Figure 5a), which is consistent with the literature [25,26]. Table 1 demonstrates the HGR and yield using different 2D Co-B NS@1D TiO₂ NFs catalyst loads. Also, the reaction time decreases with the increased catalyst amount. When the catalyst loading increased from 5 mg to 20 mg, the reaction time decreased from 12 to 3.3 min. H₂ generation tends to be rate-limited by the amount of the catalyst [25,26]. Figure 5b shows a logarithmic relationship between the reaction rate and concentration of catalyst, demonstrating that the dehydrogenation of SBH in the presence of 2D Co-B NS@1D TiO₂ NFs follows first-order reaction kinetics with respect to the catalyst concentration.

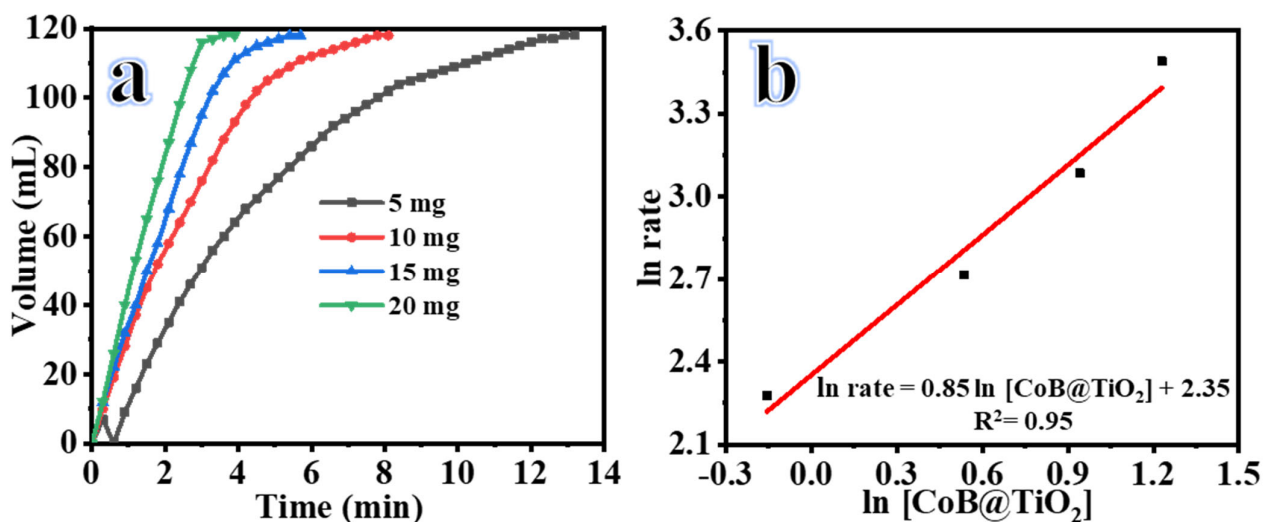


Figure 5. (a) H₂ volume vs. time for dehydrogenation of SBH at different 2D Co-B NS@1D TiO₂ NFs loading and (b) the corresponding kinetics.

Table 1. Influence of catalyst loading on the dehydrogenation of SBH.

	Catalyst Loading (mg)			
	5	10	15	20
Volume, mL	118	118	118	118
Yield, %	98.3	98.3	98.3	98.3
Reaction time, min	12	7.5	4.8	3.3
Reaction Rate, mL min ^{−1}	9.8	15.1	21.9	32.8

2.2. Influence of SBH Concentration

The effect of changing the initial SBH concentration from 26 to 105 mM on the HGR was examined while maintaining the other parameters to be constant (5 mg of catalyst, 25 °C, and 1000 rpm) throughout the experiments (Figure 6a). As the volume of H₂ generation increased with increasing SBH concentration at almost identical HGR, it can be concluded that the dehydrogenation of SBH in the presence of the 2D Co-B NS@1D TiO₂ NFs catalyst was independent of the SBH concentration and followed zero-order reaction kinetics with respect to the SBH concentration (Figure 6b).

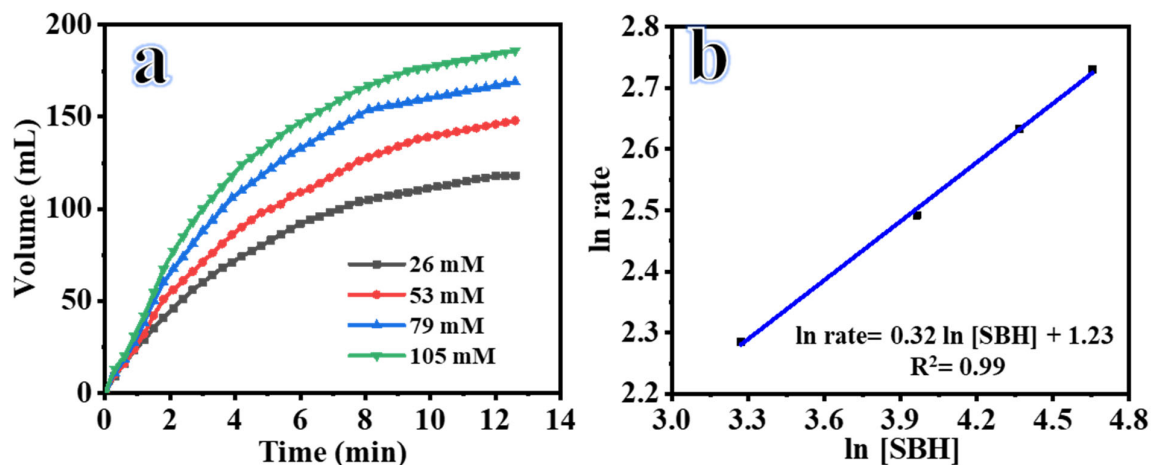


Figure 6. (a) H₂ volume vs. time for the dehydrogenation of SBH at different SBH concentrations and (b) the corresponding kinetics.

2.3. Influence of Reaction Temperature

As expected, raising the reaction temperature accelerates the reaction rate of H₂ (Figure 7). Figure 7a displays the H₂ production volume employing 2D Co-B NS@1D TiO₂ NFs catalyst at various temperatures. With an increase in temperature, the reaction rate increases rapidly. Table 2 demonstrates the HGR and yield at various reaction temperatures. The reaction time decreased for producing stoichiometric hydrogen with the increase in the reaction temperature. A plausible explanation to this observation is the ever-increasing mobility of SBH molecules, which leads to a greater number of realistic collisions beyond the energy threshold barrier [26]. Based on these observations, the reaction turned out to be zero order since the volume of H₂ production grows linearly with the reaction time. The rate of a zero-order reaction is given by the following expression:

$$r = k_0 \exp\left(\frac{-E_{act}}{RT}\right) = k_{NaBH_4} \quad (1)$$

Table 2. Influence of reaction temperature on the dehydrogenation of SBH.

	Catalyst Loading (mg)			
	298	308	318	328
Volume, mL	118	118	118	118
Yield, %	98.3	98.3	98.3	98.3
Reaction time, min	13.2	12	6	4.5
Reaction Rate, mL min ^{−1}	8.94	9.8	19.7	26.2

Plotting ln k against 1/T yielded the activation energy (E_a) to be 30.87 kJ mol^{−1} of the hydrolysis reaction process (Figure 7b). This value was at par with the others that had been achieved and published in the literature (Table 3). Also, plotting ln k_D against 1/T yielded the entropy (ΔS[#]) and enthalpy (ΔH[#]) of the activations of the hydrolysis reaction as 0.065 kJ mol^{−1} K^{−1} and 28.28 kJ mol^{−1}, respectively. The ΔH[#] and ΔS[#] values could now be used to obtain the Gibbs energy of activation (ΔG[#]) using Equation (2)

$$\Delta G^\# = \Delta H^\# - T\Delta S^\# \quad (2)$$

$$\Delta G^\# = 28.28 - 0.065T \quad (3)$$

Table 3. Performance comparison of our catalyst with those reported in the literature.

Catalyst	Conditions	E _a , kJ mol ^{−1}	Ref.
Co@ HAP	819 mg cat, 150 mM SBH, 10 wt% NaOH	53	[27]
Co-B	20 mg cat, 20 wt% SBH, 5 wt% NaOH, 20 °C	64.87	[28]
Co-Co ₂ B	10 mg cat, 0.019 g SBH, 0.025 M NaOH, 30 °C	35.245	[29]
Co-B/Cu	4 × 4 cm ² cat, 1 wt% SBH, 5 wt% NaBH ₄ , 25 °C	43.3	[30]
Co-B/Ni	1 × 1 cm ² cat, 15 wt% SBH, 5 wt% NaOH, 30 °C	94.5	[31]
Co-B/C. Vulgaris	100 mg cat, 0.66 M SBH, 1% NaOH, 25 °C	25.22	[32]
CoB/Ag-TiO ₂	10 mg cat, 1 wt% SBH, 5 wt% NaOH, 25 °C	44	[33]
CoB-MWCNT	100 mg cat, 5 wt% SBH, 5 wt% NaOH, 25 °C	23.5	[34]
Co-B/GO	50 mg cat, 5 wt% SBH, 5 wt% NaOH, 30 °C	26.2	[35]
2D Co-B NS@1D TiO ₂ NFs	5 mg cat, 26 mM SBH, 25 °C, 1000 rpm	30.87	This Work

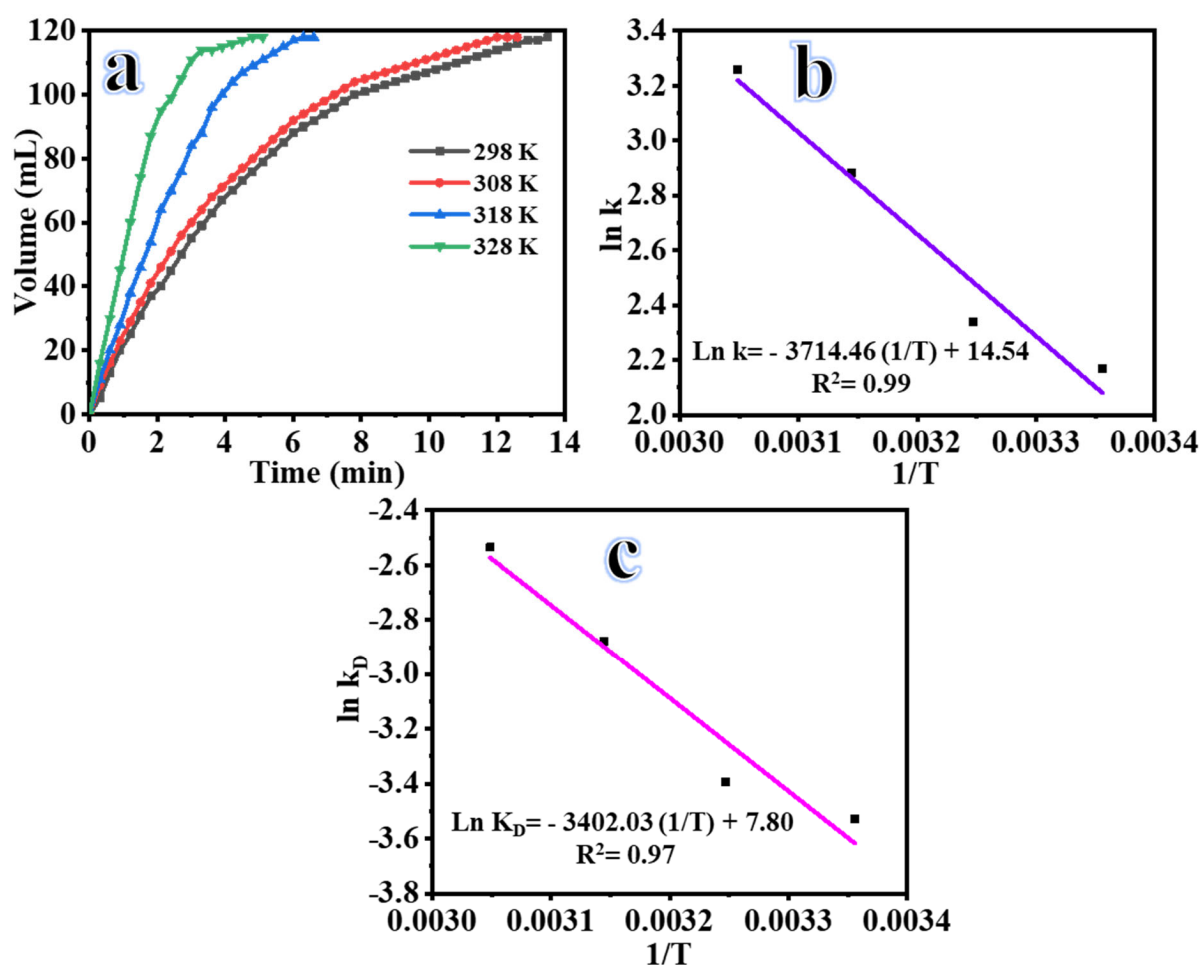


Figure 7. (a) H_2 volume vs. time for the dehydrogenation of SBH at different reaction temperatures, (b) $\ln(k, \text{rate constant})$ vs. temperature inverse, and (c) $\ln K_D$ vs. temperature inverse.

2.4. 2D Co-B NS@1D TiO_2 NFs Recyclability

For any practical application, the potential to recycle the catalyst is very crucial. The depreciation that occurs in the hydrogen production rate (Yield and HGR) with the 2D Co-B NS@1D TiO_2 NFs catalyst after going through 10 reuse cycles is depicted in Figure 8. After five repetitions, the yield efficiency of the 2D Co-B NS@1D TiO_2 NFs catalyst was compared with the intra-zeolite Co(0) nanoclusters, which preserved 59% of their activities after the 5th cycle [36], and the cobalt (Co)-based thin-film catalysts preserved 14% of their activities after the 5th cycle [37]. However, after five cycles, the depreciated activity of the Co-B/Ag- TiO_2 was slightly lower than 70% [33], 64% for the Co-B-supported on N_2 -doped carbon [26], and 37.5% for Co-B-supported carbon-derived bacterial cellulose [38]. The loss of cobalt was mostly responsible for the reduction in the H_2 yield [26]. After every run, the catalyst was recovered and cleaned extensively with water to remove residuals from the catalyst's surface, resulting in the loss of some catalyst. The formation of by-products, such as $NaBO_2$ and B_2O_3 , is another reason for the reduction in H_2 yield [38]. In addition, the surface oxidation of the Co-B catalyst is also a factor that contributes to the deactivation of the catalyst [26]. However, when the low price of the raw materials and the simple synthesis process of our catalyst are taken into consideration, these disadvantages become insignificant up to some extent.

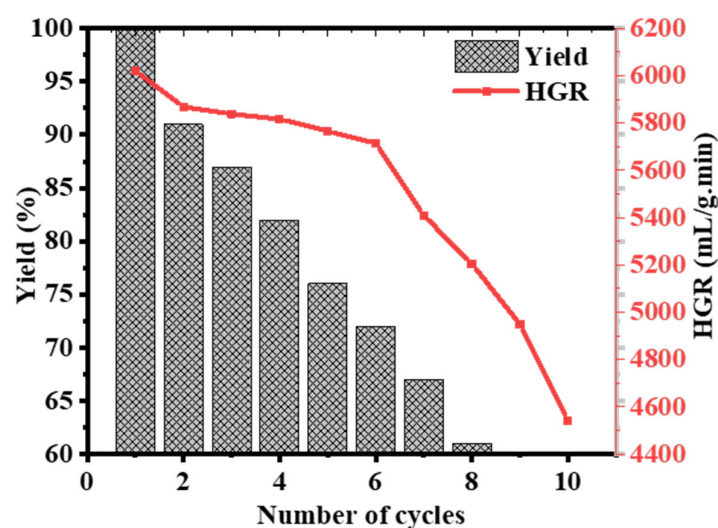


Figure 8. Recyclability of 2D Co-B NS@1D TiO₂ NFs on the dehydrogenation of SBH.

3. Experimental

3.1. Preparation of 2D Co-B NS-Decorated 1D TiO₂ NFs

First, we prepared a TiO₂ sol by mixing 2.25 g of Polyvinylpyrrolidone (PVP, average Mw 1,300,000, 98%, Sigma Aldrich, St. Louis, MO, USA) with 8 g of acetic acid (99.5%, Beijing chemical works, Beijing, China) and 8 g of ethanol (99.5%, Beijing Chemical Works, Beijing, China). The solution was then mixed with 1.5 g of titanium isopropoxide (TIIP, 97%, Sigma Aldrich, St. Louis, MO, USA). The solution was stirred continuously overnight. After the precursor solution was stirred thoroughly at room temperature to achieve the homogenous, clear, and light-yellow solution that was needed to perform electrospinning process, the solution was loaded into a plastic syringe. The voltage applied to the syringe was 18 kV, and the tip of the syringe and the paraffin sheet collector were 16 cm apart. After this, the collected mats were separated away from the sheet and calcined at a temperature of 700 °C for a period of 2 h. Then, 1 g of electrospun TiO₂ NFs was placed in a 500 mL beaker containing 100 mL of deionized water and 2 g of cobalt acetate tetrahydrate (Co(CH₃COO)₂, CoAc, 98%, Sigma Aldrich, St. Louis, MO, USA) to produce a catalyst referred from now onwards as 2D@1D Co-B@TiO₂ NF. To achieve complete dissolution of the CoAc, the solution was sonicated for 30 min. Later, an aqueous solution of sodium borohydride (SBH, NaBH₄ (98%), Sigma Aldrich, St. Louis, MO, USA) was introduced in drops to the aforementioned solution while keeping the molar ratio of Co and B to be 1:5 for the entire reductive reaction of CoAc. As soon as SBH was attached with the solution, a dark gray precipitate was formed. The as-prepared product was then filtered and washed with deionized water and absolute ethanol. The resulting product was then vacuum dried at 60 °C overnight. Under identical conditions, unsupported Co-B NPs were synthesized.

3.2. Characterization

Field-emission scanning electron microscopy (FESEM, Hitachi S-7400, Tokyo, Japan) was used to examine the surface morphology of the NFs that had been prepared. By the use of X-ray diffraction (Rigaku Co., Japan), we determined the crystalline structures of the catalysts as well as their crystal sizes ($\lambda = 1.54056 \text{ \AA}$). An investigation using X-ray photoelectron spectroscopy (XPS, AXIS-NOVA, Kratos Analytical, Manchester, UK) was carried out with the following measurement parameters: a starting pressure of 6.5×10^{-9} Torr, a resolution (pass energy) of 20 eV, and a scan step of 0.05 eV/step.

3.3. Dehydrogenation of SBH Using 2D@1D Co-B@TiO₂ NFs

The SBH solution and the catalyst were kept in a round-bottomed flask with two necks; one was sealed with a stopper, and the other was connected to a gas burette. This apparatus

was submerged in a water bath to control the reaction temperature. A thermocouple was employed to control the reaction temperature. The reactions initiated when 26 mM of alkaline aqueous solution of SBH and 5 mg of catalyst were added to a flask and stirred magnetically at 1000 rpm and 25 °C. Using the water-displacement method, the volume of gas was determined employing a gas burette at NTP. Time was recorded against the amount of evolved hydrogen. The procedure was terminated when hydrogen gas production ceased. For the sake of comparison, we also conducted the same procedure without the inclusion of any catalytic substance. All synthesized catalysts had undergone identical testing. The kinetics of SBH hydrolysis was further investigated by running the process at several concentrations of catalyst, SBH, and temperatures from 298 to 313 K. The synthesized NFs were also tested for recycle efficiency. This procedure was conducted several times using the same set of catalytic NFs to evaluate the catalyst's stability. Once the hydrolysis was complete, the catalyst was rinsed with de-ionized water, filtered, and dried under vacuum. A new 26 mM SBH was added to the regenerated catalyst in the reactor.

4. Conclusions

In this study, a 2D Co-B NS@1D TiO₂ NFs catalyst was synthesized employing electrospinning and chemical reduction methods. In terms of catalytic activity, the 2D Co-B NS@1D TiO₂ NFs performed well during the dehydrogenation of SBH. At 25 °C, a high HGR of 6020 mL min⁻¹ g⁻¹ was obtained. The kinetic analysis of the dehydrogenation process by the 2D Co-B NS@1D TiO₂ NFs catalyst showed the reaction order to be of zero order and first order with respect to the concentration of the SBH and catalyst, respectively. The activation energy E_a for the hydrolysis process was found to be 30.87 kJ mol⁻¹, whereas the ΔH and ΔS were calculated to be 28.28 kJ mol⁻¹ and 0.065 kJ mol⁻¹ K⁻¹, respectively. We demonstrated that the 2D Co-B NS@1D TiO₂ NFs catalyst is a promising candidate for the H₂ production via the selective catalytic dehydrogenation of SBH process because of its ease of fabrication, cheap price of the precursor, and high catalytic activities.

Author Contributions: Conceptualization, A.A. and A.Y.; methodology, A.Y. and I.Y.Q.; validation, M.M.E.-H. and S.M.M.; formal analysis, I.M.M. and A.S.M.A.; investigation, N.Z. and I.Y.Q.; resources, A.A. and S.M.M.; data curation, M.M.E.-H. and A.S.M.A.; writing—original draft preparation, I.M.M., N.Z., A.A. and M.M.E.-H.; writing—review and editing, A.Y., I.Y.Q., S.M.M. and A.S.M.A.; visualization, N.Z., I.Y.Q., S.M.M. and A.S.M.A.; supervision, I.M.M., A.A., A.Y. and M.M.E.-H.; project administration, I.M.M., N.Z., A.A., A.Y., I.Y.Q., S.M.M., A.S.M.A. and M.M.E.-H.; funding acquisition, I.M.M. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data presented in this study are available from the corresponding authors upon reasonable request.

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Conflicts of Interest: The authors declare no conflict of interest.

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