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# Spill Behaviour of Hydrogen Carriers as Alternative Fuels for Ships

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**Abstract** – Hydrogen carriers, such as liquid organic hydrogen carriers (LOHCs) and borohydrides, are promising zero-emission alternative fuels for ships. Bringing these hydrogen carriers on board, however, creates new challenges. A major challenge is their spill behaviour. Knowing the spill behaviour is paramount to avoid large-scale environmental disasters. This paper investigates the spill behaviour of four hydrogen carriers (and their conjugates): sodium borohydride, ammonia borane, dibenzyltoluene, and n-ethylcarbazole. The hydrogen carriers were all dissolved in artificial seawater to test their behaviour. Sodium borohydride reacts with seawater, as it also reacts with pure water. However, contrary to expectations, it reacts faster with seawater than regular water. The reaction mechanism behind this is unknown. Ammonia borane does not visibly react with normal water or with seawater. Dibenzyltoluene sinks and forms tiny bubbles which are easily perturbed. Unfortunately, perhydro dibenzyltoluene could not be tested due to technical problems. N-ethylcarbazole breaks up into smaller pieces and predominantly stays afloat, likely due to the surface tension of water. Perhydro n-ethylcarbazole floats but is barely visible in seawater due to its transparency. Preventive measures must be established to avoid large-scale spills if these substances are utilised on ships, as they are likely challenging to clean up.

**Keywords:** hydrogen carriers; alternative fuels; spill behaviour; water pollution; LOHC

## 1. Introduction

The International Maritime Organization (IMO) aims for ships to achieve net Greenhouse Gas (GHG) emission neutrality by 2050 [1]. Adopting alternative fuels is vital in reaching this target, but conventional options like ammonia, methanol and hydrogen have significant drawbacks, such as toxicity, storage challenges or safety concerns [2]. A promising solution lies in using hydrogen carriers, which can be non-toxic, non-explosive and offer high energy densities. Previous research identified four promising hydrogen carriers [2]: two liquid organic hydrogen carriers (LOHCs), dibenzyltoluene (DBT) and n-ethylcarbazole (NEC), and two boron-based carriers, sodium borohydride (SB) and ammonia borane (AB). These four carriers have relatively high energy densities, over 6.9 MJ/kg for the LOHCs and over 20 MJ/kg for AB and SB. Furthermore, they release hydrogen on demand, can be regenerated and are liquid (LOHC) or solid (SB and AB) [2]. The empty, spent fuels must be stored on board as well and can be discharged and regenerated at a shore facility.

Using these hydrogen carriers as an alternative fuel on ships will come with additional challenges compared to land usage, including space and weight limitations and safety concerns. Currently, post-Panamax ships can use up to 20,000 m<sup>3</sup> of fuel, and Panamax container ships and large cruise ships can use up to 7500 m<sup>3</sup> [3]. Even an inland tank barge (90m long) can carry up to 4500m<sup>3</sup> [3]. When using hydrogen carriers, these tanks can be up to 4 times larger to accommodate the same energy storage [2]. The potential for fuel spills is thus significant, and the use of chemicals as fuels shifts the risk from relatively small-scale contamination to the potential for large-scale, unpredictable spills, which can cause catastrophic damage [4]. For instance, the Prestige oil spill resulted in 66% less species richness in specific locations, and these types of effects can persist up to 40 years after the incident [4,5]. Moreover, such spills can have significant socio-economic effects. Unlike oil spills, the behaviour and impact of chemical spills are poorly understood and little documented. Additionally, maritime spills especially have the issue of social amplification, resulting in a skewed public view of marine pollution issues [4].

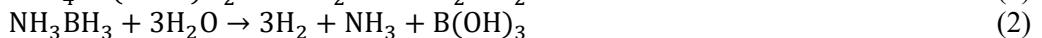
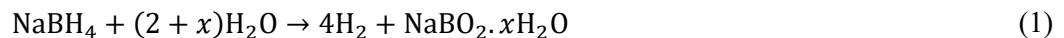
Critical parameters such as the predicted no-effect concentration (PNEC) and the GESAMP (Group of Experts on the Scientific Aspects of Marine Environmental Protection) hazard rating and persistence, bioaccumulation, and acute toxicity (PBT) are essential for evaluating spill hazards. However, these ratings do not predict how a substance behaves during a spill. Despite the OPRC-HNS protocol of the IMO (in force) [6] and the HNS convention (not in force) [7], the field of

chemical spill treatment at sea remains underdeveloped, as highlighted in a white paper that discusses the dangers of hazardous and noxious substances (HNS) spills and compares them to oil spills [8].

Understanding the behaviour of the spilt chemical in the sea is of utmost importance, yet it remains to be discovered for all four above-mentioned hydrogen carriers. Existing classifications are usually based on laboratory data, in which factors in the maritime environment, such as wind and sun, as well as the salinity of seawater, are not considered [9]. Notably, *in situ* results incorporating environmental influences can differ significantly from laboratory outcomes [9]. This paper aims to address this gap by performing tests in seawater, despite still being in a lab setting, to offer a more accurate understanding of the behaviour of these hydrogen carriers under more realistic maritime conditions.

### 1.1. Background

The hydrogen carriers are split up into two types. The first group comprises sodium borohydride (SB,  $\text{NaBH}_4$ ) and ammonia borane (AB,  $\text{NH}_3\text{BH}_3$ ). These compounds are generally soluble in water, ranging from 30 to 55 g per 100 mL [10,11]. However, they are also likely to react with water, as can be seen in equations 1 (SB) [12] and 2 (AB) [11]. During these reactions, hydrogen is released. In the case of AB, ammonia is also produced.



The second type of hydrogen carrier investigated in this study are liquid organic hydrogen carriers (LOHCs). These carriers are oil-like and have low vapour pressures. The latter means they are not likely to evaporate. Furthermore, LOHCs generally exhibit low solubility in water and are persistent [13,14]. For instance, dibenzyltoluene is thought to be only soluble in the range of ng/L, and N-ethylcarbazole has a solubility of several mg/L [13]. LOHCs release hydrogen through an endothermic reaction, as shown in equation 3, requiring significant amounts of heat at elevated (higher than 200 C) temperatures. Consequently, it is doubtful that these hydrogen carriers would release hydrogen during a spill in (sea)water.

### 2. Method

This research involved several experiments presented in separate sections due to the different approaches required for the borohydrides (AB and SB) compared to the LOHCs. As literature findings sometimes diverge from experimental results, as noted by Le Floch et al. [9], this study will also mention possible discrepancies.

The experiments focussed on the solubility and behaviour of the mentioned hydrogen carriers (both hydrogenated and dehydrogenated) in seawater. Seawater is synthesised using the Instant Ocean brand Seawater from Aquarium Systems, an ion mixture. The mixture contains sodium-, chloride-, sulfate-, magnesium-, calcium-, and carbonate- ions, among others. Table 1 gives an overview of the substances used, the suppliers, and the purity grades.

Table 1: Used substances, suppliers and grades

Substance	Grade	Supplier
N-ethylcarbazole	99%	Fisher Scientific
Perhydro n-ethylcarbazole	>95%	Voyex, SH2IPDRIVE partner [17]
Dibenzyltoluene	99%	Biosynth
Perhydro dibenzyltoluene	-	Not Available
Sodium borohydride	98%	CPH Chemicals
Ammonia borane	>85%	TCI Chemicals
Instant Ocean Seawater	-	Aquarium Systems

## 2.1. Borohydride Method

The borohydrides (SB and AB) were tested for their reaction with seawater (see eqs. (1) and (2)). A pneumatic through through is used to measure the hydrogen production from both SB and AB when reacting with seawater. Figure (1) gives an overview of the test setup used in this research. The resulting substance from the SB reaction (visible in equation 1),  $\text{NaBO}_2$ , will dissociate in water, resulting in  $\text{Na}^+$  and  $\text{B}(\text{OH})_4^-$ -ions. These strongly alkaline ions can result in a strongly alkaline solution [15]. To avoid forming such a very strongly alkaline solution, to mimic the conditions of open seawater, and because of limitations due to safety in the test set-up, a total of 0.15g of SB was added to a large surplus of 600mL of water. In the case of complete conversion of all SB, this is predicted to result in a pH of 10.52, regardless of whether salt is added. AB reacts similarly, raising the pH level of the solution to 9.1 when added to water [16], and thus the same amount of water was used. Experiments involved placing 0.15g of SB or AB in an airtight Erlenmeyer flask filled with seawater. This flask is connected via a tube to a pneumatic trough, allowing us to measure the volume of hydrogen gas the reaction generates. Temperature conditions were adjusted using a heating plate when necessary.

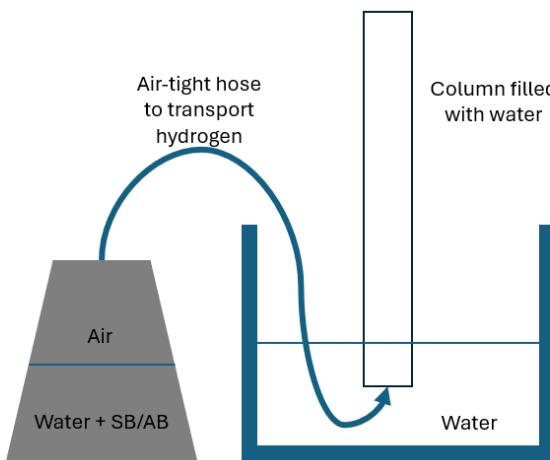


Fig. 1: Schematic overview of test set-up to test self-hydrolysis of ammonia borane and sodium borohydride in (sea)water.

## 2.2. LOHC Method

The behaviour of both LOHCs was tested. Since hydrogenated LOHCs are not commercially available, NEC was synthesised by SH2IPDRIVE partners [17], with only small amounts available for testing. Perhydro dibenzyltoluene could not be synthesised on time due to the failure of the machinery required to hydrogenate dibenzyltoluene. A small beaker containing instant ocean artificial seawater at a temperature of 22 °C was used for behaviour tests. The LOHC was added to the beaker, with LOHC always in a significantly lower quantity than seawater. The main aim is to categorise these substances using the Standard European Behaviour Classification (SEBC) [9]. The SEBC classifies substances into categories such as floaters, sinkers, and evaporators. These main groups have subgroups, e.g. substances that dissolve or evaporate. Combined, the SEBC has 12 behaviour groups [9].

## 3. Results and Discussion

As the substances behave in different ways, again, two different sections are required. First, the results of the borohydride tests will be shown, followed by the results of the LOHC tests.

### 3.1. Sodium borohydride

Figure 2 presents an overview of the SB reaction results. The data indicate that seawater, compared to pure water, significantly enhances the hydrogen production rate. Regardless of the temperature, the seawater mixture consistently accelerates hydrogen generation compared to pure water. Due to the very low reaction rates at low temperatures, the tests could not be done at lower temperatures. However, estimates can be made for these lower temperatures based on the

Arrhenius rate equation. The results from Figure 2 indicate unexpected behaviour. The half-life time for self-hydrolysis of SB has been reported to follow equation (4) [18]:

$$\log\left(\frac{t_1}{2}\right) = \text{pH} - (0.034 T - 1.92) \quad (4)$$

with the temperature T in Kelvin. This equation indicates a slower reaction rate for a higher pH value. As the starting pH value of seawater (8.2) is already above that of pure water (7.0), it can be expected from equation (4) that the SB hydrolysis reaction should be slower in seawater than in pure water. However, the results indicate the opposite: the reaction rate is faster in seawater than in pure water. Both pH levels increased, as expected, but they increased to a very similar level of around 10.5. As both rose to this similar level, it can be safely assumed that the pH change does not explain the observed change in reaction rate. Something in the sea salt enhances the reaction rate, but further research is needed to determine the mechanism driving this increase.

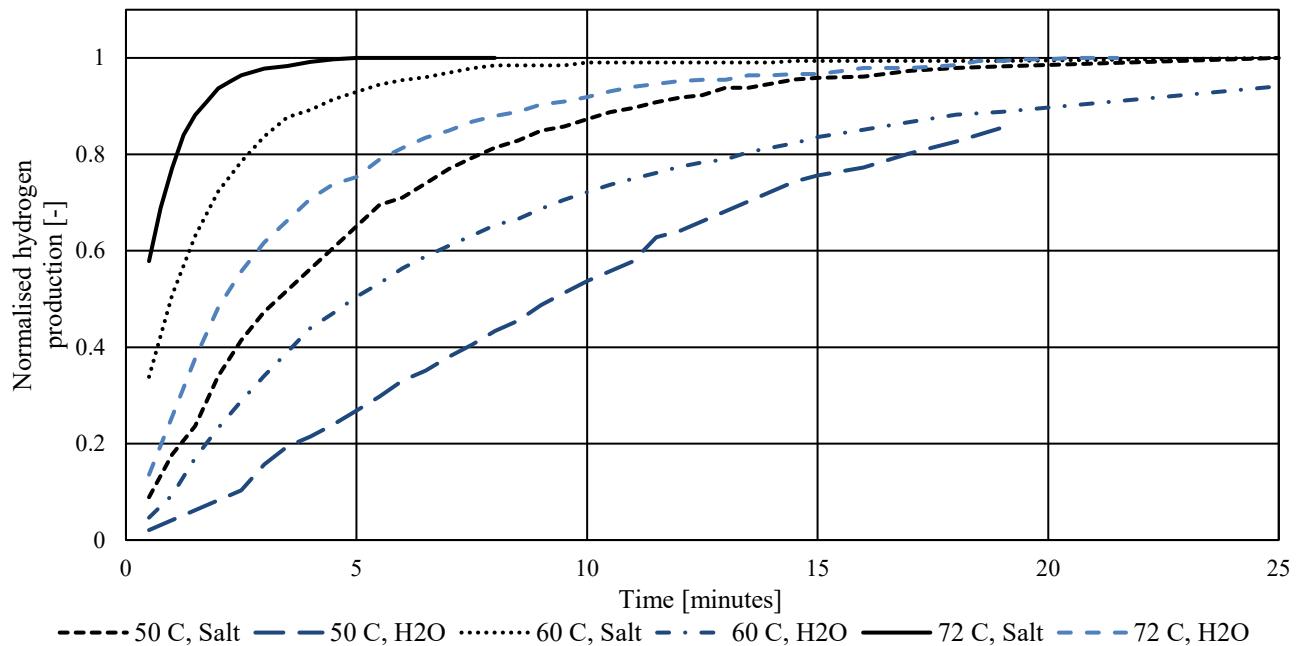


Fig. 2: Results of self-hydrolysis of sodium borohydride in (sea)water. Hydrogen production as function of temperature and water type

### 3.2. Ammonia Borane

Ammonia borane's self-hydrolysis in water is reportedly slow, showing no reaction after extended periods of up to 80 days [16]. Adding (solid) acids will lead to fast hydrogen release [16].

In seawater, with a pH of about 8.2, ammonia borane did not react within the time frame of an hour. As the reaction mechanism is likely similar to SB but with a much lower rate, the test was stopped as it was unlikely to react further on the time scale of hours.

Because AB can be stable for up to 80 days [16], it is likely to disperse before it starts reacting, and it may not react at all. Since AB does not dissociate, it will not release ammonia or other harmful substances into the environment. However, dissolving AB in water may elevate the local pH to 9.1, depending on the concentration [16].

### 3.3. LOHCs

Figure 3 shows the (theoretical) densities of LOHCs compared to seawater as a function of the temperature [19,20,21]. It indicates that at typical seawater temperatures (0 °C - 30 °C), both dehydrogenated versions (H0DBT and H0NEC) are denser than seawater, and both hydrogenated versions (H18DBT and H12NEC) are less dense than

seawater. The dehydrogenated version of NEC is solid until approximately 68 to 70°C, and its exact density as a function of temperature in a solid state is unknown to these authors. Table 2 gives an overview of the behaviour of these substances in seawater, and Figure 4 shows pictures of the results.

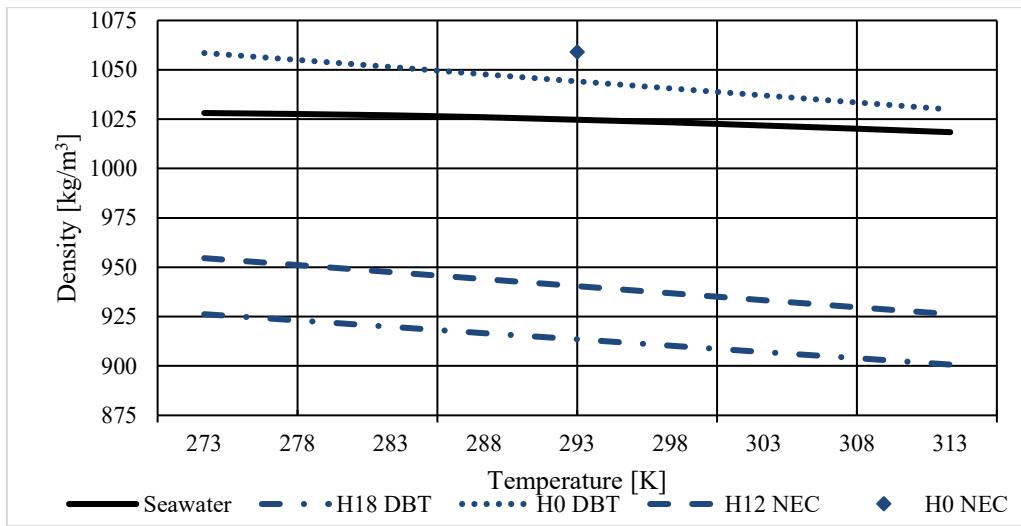


Fig. 3 Theoretical densities of LOHCs and seawater for a reasonable range of seawater temperatures

N-ethylcarbazole first aggregates on the surface of the water, but upon movement, it disintegrates into individual fragments. These fragments are visible in Figure 4a. Some of these fragments settle to the bottom, while the majority remains buoyant. The structure consists of elongated particles resembling shards or splinters. Upon agitation, the fragments on the bottom are resuspended and dispersed, following the motion. The buoyancy of NEC is unexpected as, according to Fig. 3, the density of NEC is higher than that of seawater. The fragments remained buoyant even after vigorous agitation. However, the particles immediately drop to the bottom after adding dish soap to remove surface tension. Therefore, surface tension likely keeps NEC afloat, and NEC is a sinker. Testing in demineralised water revealed notable differences. In seawater, both large and small particles settled to the bottom, whereas in demineralised water, only smaller particles did. Additionally, NEC settled more quickly after movement.

Perhydro n-ethylcarbazole (H12-NEC) can be seen in Figure 4b. H12-NEC is barely visible in seawater, as it is transparent. Its visibility improved slightly after adding a dye (indigo carmine from Sigma Aldrich). However, it was sufficiently visible, even without dye, to allow some observations. Notably, H12-NEC remains afloat as expected. It forms extremely small droplets. Figure 4b illustrates some droplets floating in the seawater, away from the walls. H12-NEC disperses readily and appears to form a thin film on top of the water if undisturbed. Upon agitation, the tiny droplets are formed. When adding dish soap, H12-NEC does not sink, but the film on the water's surface becomes more pronounced.

Dibenzyltoluene forms large bubbles ranging from several millimetres to approximately a centimetre. As expected, most of these bubbles settle to the bottom, as visible in Figure 4c. Although the bubbles encounter each other, they do not coalesce. There appears to be a maximum bubble size of approximately 1 cm. A single large bubble, around 1 cm, remains at the surface. After agitation, the bubbles break apart and become significantly smaller. The surface bubble increases in size, reaching approximately 2 centimetres. An additional large bubble forms around the stirring magnet. With more vigorous agitation, the bubbles reduce further, with a maximum diameter of a few millimetres. However, when the system is at rest again, the bubbles grow larger due to coalescence. The surface bubble returns to its original size of about 1 cm. Notably, the bubble around the stirring magnet persists and cannot be displaced, even with substantial movement of the magnet. Due to the low solubility and low vapour pressure, dibenzyltoluene can be categorised as a 'sinker' in the SEBC classification.



Fig. 4 Behaviour of LOHCs in artificial seawater

As perhydro dibenzyltoluene could not be synthesised on time, no results can be displayed in this paper, and the behaviour of dibenzyltoluene is left for further research. However, it is still recommended that perhydro dibenzyltoluene be tested.

### 3.4. Discussion

The enhanced hydrogen production rate of SB in seawater relative to pure water may cause potentially dangerous situations at sea. However, it must be realised that the increase in reaction rate is relatively mild, i.e. not by orders of magnitude. If the produced hydrogen gas is dispersed fast enough, it may not ignite. However, hydrogen explosions cannot be ruled out in the presence of an ignition source, such as a fire, and at relatively high sea surface temperatures.

Table 2: Behaviour of LOHC substances in seawater

Substance	Description of behaviour	Classification
N-ethylcarbazole	Predominantly floats. The structure is analogous to shards, where individual fragments do not remain cohesive. Some fragments either sink or circulate within the liquid. As the floating is due to surface tension, NEC is considered a sinker, but it can also behave as a floater in practical surroundings.	S (Sink), but depending on the particle size, can also be F (float)
Perhydro n-ethylcarbazole	Floats, forms tiny droplets when disturbed. It appears to form a layer when undisturbed, but it is difficult to conclude due to the bad visibility of perhydro n-ethylcarbazole in water.	F (Float)
Dibenzyltoluene	Predominantly sinks. Forms bubbles ranging in size from millimetres to approximately 2 centimetres. A single bubble remains buoyant. Upon disturbance, bubbles undergo fragmentation, reducing in size to only millimetres. However, over time, these small bubbles coalesce again into larger structures. Maximum bubble size appears to be ~2 centimetres.	S (Sink)

Ammonia borane appears to be relatively safe; however, its impact is highly dependent on the amount released and the specific spill location, as it has the potential to raise the pH of the surrounding environment. In cases where a substantial amount is released, the pH of the surrounding seawater may increase locally, which could be detrimental to certain sensitive species. However, the immediate impact will likely be negligible if the spill occurs in the open ocean.

The results indicate that all three tested LOHCs are unlikely to be easily cleaned up due to their dispersion. The tendency to fragment during movement suggests that these substances would likely become even more dispersed in a real-world marine environment where wind and waves are present. Even though NEC is officially classified as a sinker, it is likely to stay afloat during an actual spill because it is very hydrophobic, and its surface tension is sufficient to keep small particles afloat. Whether it floats or sinks depends on the particle size, as larger objects have a larger volume-to-surface ratio and are, therefore, more likely to sink. The poor visibility of H12-NEC in seawater suggests that locating and cleaning it after a spill would be highly challenging. DBT tends to sink, so it poses a different cleanup challenge than floating substances. Moreover, its tendency to adhere to surfaces will complicate the cleanup process. Therefore, effective cleanup of all these substances in the event of a spill may prove highly challenging.

#### 4. Conclusion

This study aimed to evaluate the behaviour of four hydrogen carriers in seawater to assess their potential spill impact. The findings indicate that SB does not behave as expected, reacting more rapidly in seawater than in pure water. The associated risks of this behaviour depend on seawater temperature and the quantity of SB released, meaning that hydrogen gas can be released in the seawater, escaping to open air. The scale and impact require further research. In contrast, AB performed as anticipated, showing no response in the seawater environment, suggesting minimal immediate risk in case of a spill.

For the LOHCs, NEC did not directly behave as expected, as it predominantly floated in the seawater, likely due to surface tension. Pieces of NEC did circulate and sink, with more pieces sinking in seawater than in demineralised water. When dish soap was added, and the surface tension was broken, all particles dropped to the bottom. The behaviour of NEC depends on particle size, with smaller particles likely floating and larger ones sinking.

Perhydro n-ethylcarbazole (H12-NEC) floats in seawater, as expected. However, it disperses quickly and is barely visible in seawater due to its clarity. The poor visibility is likely to complicate the clean-up process.

DBT conformed to expectations, as it predominantly sank. Additionally, it formed small bubbles which were easily perturbed. This behaviour suggests it could be very challenging to clean up. Unfortunately, due to material limitations, perhydro dibenzyltoluene remains untested. Thus, in the case of LOHCs, developing preliminary cleanup strategies in advance is essential. DBT, for example, is toxic and persistent and may be complicated to remove once released into the marine environment.

Overall, these findings highlight the importance of understanding the environmental behaviour of hydrogen carriers before their widespread use in maritime applications. Given the large size of fuel tanks (up to 20000 m<sup>3</sup>), a spill of such magnitude could have severe environmental implications, with possible extensive and long-lasting impact on marine ecosystems, particularly as these substances appear challenging to clean up. Therefore, if these substances are to be used on ships, it is imperative to establish preventive measures to avoid large-scale spills.

#### Acknowledgements

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