Borophenes to borophites: exploration through electron counting

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Isoelectronic comparison to graphene and magnesium boride (MgB_2) explains the inevitable holes in borophenes. Similar qualitative analysis of stacking borophene layers towards an equivalent of graphite brings up several possibilities, which are presented here. Electron counting indicates that, in addition to the van der Waals interactions commonly seen in graphite, interlayer 2c–2e and multi-centre bonds, and hole density play an important role in the structure of borophites. The complexity that arises here may surpass that of 3D boron allotropes.

Keywords: Borophenes, borophites, bond density, electron count, hole density.

ALLOTROPES of boron are more complex to comprehend than most elements. In contrast to the well-defined modes of construction of allotropes of carbon, boron with only three valence electrons brings in multicentre bonding and a plethora of possibilities. After reviewing the ways to understand 3D allotropes of boron in an article in this journal more than 12 years ago, one of the present authors (E.D.J.) had concluded: 'The recent proposal of fullerene-like boron clusters, boron nanotubes and one atom thick boron layers are evidences for the use of an understanding of the structure and bonding using electron counting rules. Chemistry of boron, boranes, macropolyhedral boranes, elemental boron and boron-rich solids form an unknown continent to be explored and understood. Only the initial rules of the game have appeared.'[1] Until recently, 2D allotropes of boron were unknown. Unlike graphene, which was first peeled out of graphite[2], there was no 'borophite' available to peel-off borophene[3]. During the last six years, several 2D allotropes of boron were generated mostly by atom deposition strategies on metal templates[4–16]. The 2D allotropes of boron (borophenes) are turning out to be just as complex as its 3D allotropes. We present an exploration of stacking 2D allotropes and the possibilities it provides using electron counting, leading to potentially infinite variety of 'borophites', an acceptable name in the family of graphite, graphene and borophenes. We begin by comparing electron counting in well-known classical benzene (C_6H_6, flat hexagon, in graphene and magnesium boride (MgB_2) (Figure 1 *a–e*). The consequence of replacing each Mg by a boron to construct borophene, and the inevitability of holes in stand-alone borophenes are discussed next (Figure 1 *f–i*)[17,18]. This is followed by a discussion of the ways in which holes are controlled by external templates and by formation of bilayers with possible localized interlayer bonding[19–22]. Multilayer stacking provides greater variety by a combination of holes, interlayer bonding and traditional van der Waals interactions leading to the variety possible for 'borophites'. These possibilities are briefly compared to available experimental and computational data at the end[23].

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Graphite, graphene, MgB_2 and borophene

The electronic structure of benzene with its localized 2c–2e σ-bonds and the electron-sufficient valence bond description of π-bonds is common knowledge. Polycondensation of benzene to graphene is straightforward. The bonding in the σ-framework of graphene and the delocalized description of π-electron network retain the electron-precise nature of carbon and lead to the properties of graphene and graphite. Formal replacement of each carbon of a single-layer graphene by a B^- and for every two B^- an Mg^{+2} added as a layer of Mg^{+2} gives graphitic MgB_2 (Figure 1 *d* and *e*). The charge separation used in the electron counting between the alternating B^- and Mg^{+2} layers is far from complete and provides for the dramatic difference between graphite and MgB_2 in conductivity and other properties[24]. The electron-sufficient sheet of graphene and the corresponding MgB_2 with all-boron graphene–Mg 2D structure provides a starting point to borophene with its triangular network. Let us replace each Mg in Figure 1 *e* with a boron atom. Whether B atoms are from replacement of Mg or of the B-graphite, these are identical and now the added boron forms a part of the flat triangular network (Figure 1 *f*). This is similar to the planar and quasi-planar bare boron clusters which have a planar triangular sheet[22,25–31]. In fact, the name 'borophene' was first introduced by Piazza *et al.*[31] to describe the planar structure of B_{36}, a triangular array with a hexagonal hole (Figure 1 *g*)[31]. The structure of 2D-borophene and increase in its stability by introducing holes were studied by many researchers[18,32–34]. Assuming that the basic electronic structure remains

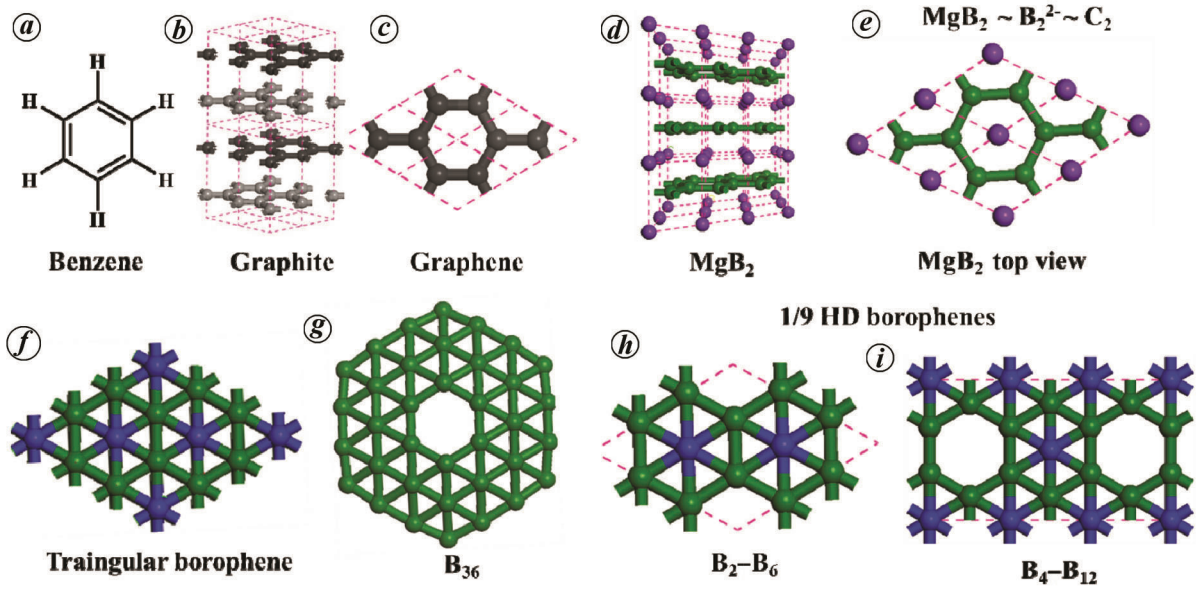

Figure 1. ***a***, Benzene showing one of a classical valence-bond resonance structure. ***b***, Graphite. ***c***, Graphene. ***d***, MgB_2. ***e***, 2D projection of B^--graphene-Mg^{+2} bilayer. ***f***, Borophene (hole density (HD), zero). ***g***, B_{36} (with a hexagonal hole). ***h***, α-Borophene (HD 1/9). ***i***, Alternative distribution of holes in HD 1/9. For clarity, the doped boron atoms are shown in blue colour.

Table 1. Binding energy (BE) and relative binding energy (RE) of different borophenes with varying hole density (HD) relative to the most stable α-borophene (1/9 HD). The energies are calculated using DFT-PBE functional[16]. The β_{15} sheet corresponds to Figure 1 *h* and α sheet to Figure 1 *g*

Borophene sheet	HD	BE (eV)	RE (meV)
δ_6 Flat	0	–5.689	312
δ_6 Puckered	0	–5.905	96
α_7	1/16 (0.062)	–5.911	90
α_8	1/12 (0.083)	–5.942	59
α_9	1/10 (0.100)	–5.969	32
α	1/9 (0.111)	–6.001	0
β_{15}	1/9 (0.111)	–5.973	28
β_5	2/15 (0.133)	–6.000	1
β_{12}	1/6 (0.167)	–5.947	54
χ_3	1/5 (0.200)	–5.958	43
δ_4	1/4 (0.250)	–5.688	313
δ_3	1/3 (0.333)	–5.101	900

similar to graphene, there is an increase in the valence electrons available when Mg is replaced by boron, an increase of one electron per each replacement (Mg–B_2 versus B–B_2). If we consider the stoichiometry Mg_3B_6, there are three extra electrons in B_3–B_6. A way of getting back to an electron count similar to graphene is to eliminate one boron, leading to B_2–B_6 instead of B_3–B_6. In other words, only eight boron atoms must remain instead of nine. The hole generated is obviously a hexagon. Thus, there should be a hexagonal hole for every nine boron atoms; the ninth atom is missing leading to a hole density (HD) of 1/9 (Figure 1 *h*). The triangular sheet with no holes corresponds to zero HD (Figure 1 *f*) and the graphene-like hexagonal honeycomb boron sheet (similar to Figure 1 *c* with all C atoms replaced by B) has 1/3 (3/9) HD. Obviously, within a given HD, the distribution of holes can be varied resulting in a variety of different structures. Figure 1 *i* gives an alternative to 1/9 HD borophene (Figure 1 *h*). Computational results on stand-alone 2D borophenes with and without holes support this analysis[17,18,32–35].

The relative energies are indicated as binding energy per boron (meV), calculated in relation to the most stable 2D allotrope with 1/9 (0.111) HD. The graphene-like sheet with hexagonal honeycomb structure (HD 1/3 (0.33)) is the least stable, 900 meV higher in energy than the most stable structure. A flat structure with zero HD is not a minimum and is higher in energy by 312 meV (Figure 1 *f*). A striated structure where alternative rows go up and down is only 96 meV higher than the best structure[32]. Increasing HD increases the binding energy till the best structure with 1/9 HD is reached. Further increase in HD decreases the binding energy (Table 1). There are several ways of distributing the holes in the 1/9 HD borophene, e.g. Figure 1 *h* and *i*. The energy differences between these alternatives are not large. Such multiple possibilities exist for sheets with other HDs and the variety possible in borophenes is obvious in comparison to the definite structure of graphene. This also shows the advantage of borophene in changing electron density. In graphene it is necessary to replace a carbon atom with say boron or nitrogen, to bring deficiency or excess of electrons. In borophene, this can be achieved by varying HD.

Additional ways of attaining the electron count–bilayer formation

The success in generating single-atom-thick borophene layers on metal surfaces gives a hint about the ways of

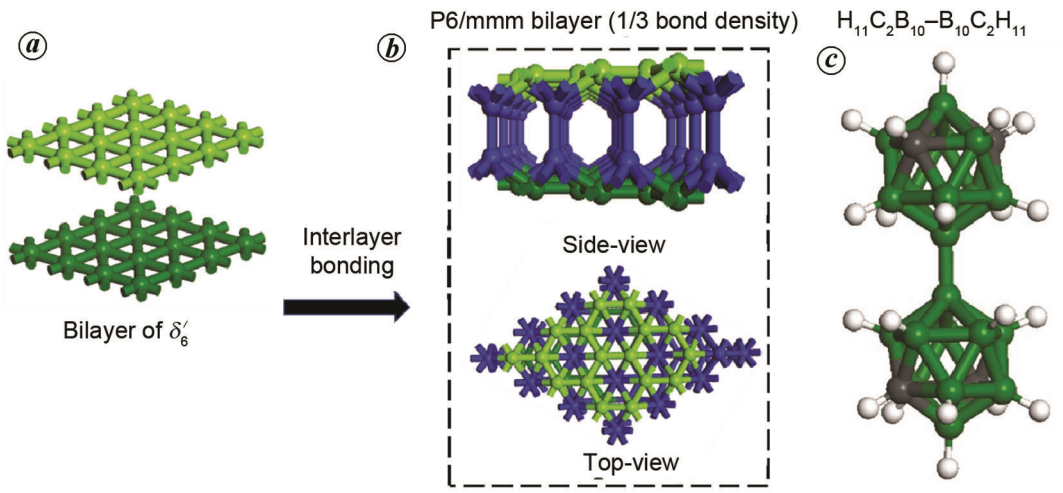

Figure 2. ***a***, Two triangular 2D monolayers with zero HD. ***b***, P6/mmm bilayer of zero-HD borophene and formation of interlayer B–B bonds. Each boron obtained by replacing Mg of Figure 1 *d* forms a 2c–2e bond with the next layer. ***c***, Dimer of the neutral carborane, $H_{11}C_2B_{10}$–$B_{10}C_2H_{11}$.

stabilizing borophenes. Available experiments indicate that the monoatomic layers have varying number of hexagonal holes depending on the metal, the boron precursor, and the rate and temperature of deposition. Hexagonal holes are an indication of the strength of the borophene–metal surface interaction; when there is electron deficiency donation of electrons from the metal stabilizes the borophene. Larger the number of holes (HD) and consequent electron requirement, higher will be the electron donation from the metal surface and consequent stabilization[5,11,12,17,18,35].

Another way of stabilizing borophenes is to form bilayers with limited interlayer bonding[19,20,22]. Let us consider two layers of zero-HD borophene by AA stacking (Figure 2 *a*), where each atom of a layer is above an equivalent one in the other. If a localized 2c–2e B–B bond is formed between two atoms one from each layer, one electron will be removed from the delocalized π-bonding system of each layer (Figure 2 *b*). The two atoms involved will move away slightly from the planar skeleton and towards each other, forming apex atoms of two hexagonal pyramids, decreasing the B–B bond length. This is not as improbable as it appears; an exohedral B–B bond in the dimer of the neutral carborane, $H_{11}C_2B_{10}$–$B_{10}C_2H_{11}$ is not far-off (Figure 2 *c*). In the bilayer, the new B–B bond stems from two hexagonal pyramids, one from each sheet. If there are three such bonds for every nine boron atoms of one layer, it is equivalent to removing one boron atom to form a hexagonal hole. Just as the HD of 1/9 brings in an electron count similar to graphene, a bond density (BD) of 1/3 (3/9) has the same effect in a zero-HD bilayer.

Depending on HD, borophene sheets can adopt varying number of interlayer bonds (BD). Let us consider an electron-rich borophene sheet with 1/12 HD, which is less than the ideal 1/9. The HD can be compared by taking the denominator as the lowest common multiple. Thus, we have 3/36 HD instead of the required 4/36: an extra boron atom or three extra electrons for every 36 atoms. If there are three interlayer bonds in the bilayer of borophene sheets of 3/36 (or 1/12) HD, the electron requirement is satisfied. What is the BD here? The total number of atoms present is 36 – 3, i.e. 33. Therefore, bond density is 3/33 = 1/11. Note that HD is calculated using the total number of atoms that would have been present had there been no holes. BD is calculated by the number of interlayer bonds within the number of atoms present (i.e. after subtracting the holes from a zero-HD sheet). Thus, a bilayer of 1/12 HD sheet needs a BD of 1/11 to keep the same electron count as a monolayer of 1/9 HD.

Multilayer borophenes and borophites

In principle, graphite-like van der Waals stacking of electron-sufficient borophene monolayer or bilayer can produce borophites. Since the α-monolayer (HD 1/9) is equivalent to graphene in terms of electron count, a van der Waals stacking of the same may resemble graphite. It is also possible to envisage infinite van der Waals stacking of bilayers and other multi-layers, which in turn are held by 2c–2e interlayer bonds as an extension of the approach seen in the zero-HD (1/3 BD) bilayer.

Let us consider the schematic representation of borophene layers (Figure 3 *a*–*d*). Suppose each layer consists of 18 B atoms of triangulene-based borophene sheet with zero HD. This has two boron atoms (six electrons) more than what is ideal in comparison to 1/9 HD. Therefore, the formation of six interlayer B–B bonds makes the bilayer (represented by blue lines in the schematic presentation as described in Figure 2 *b*) electron sufficient. Figure 3 *a* gives a schematic representation of the same. Each layer here

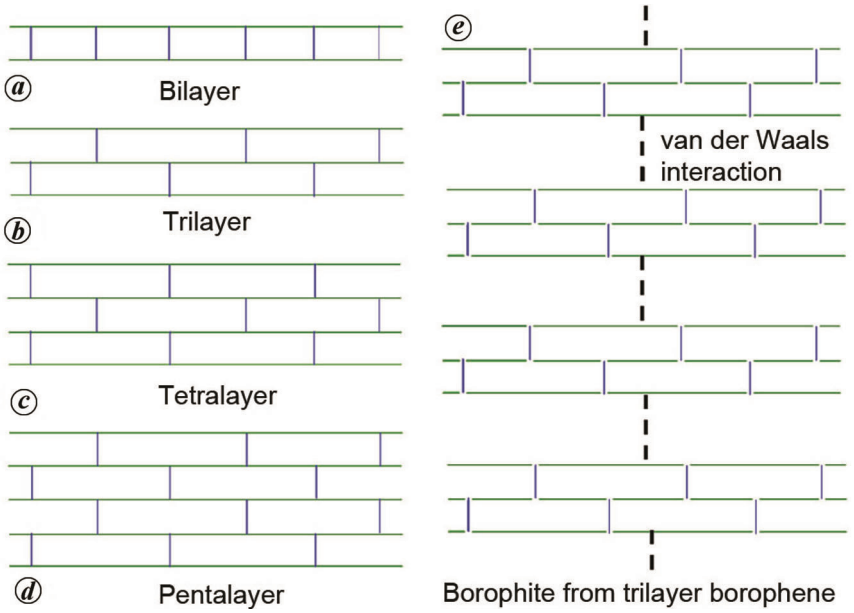

Figure 3. Schematic representation of stacking between multiple layers forming covalent 2c–2e bonds. ***a***, Schematic bilayer for Figure 2 *b* arising from zero-HD monolayer. ***b***, Trilayer – The middle layer maintains bond density (BD) of 1/3 by forming bonds with the upper and lower layers. The outside layers can adopt a HD of 1/18 to maintain the same electron count. The green lines represent borophene layers and blue lines represent the 2c–2e B–B bonds. ***c***, Tetralayer – the two inside layers maintain BD of 1/3 by forming bonds with the adjacent upper and lower layers. The two outside layers (top and bottom) adopt a HD of 1/18 to maintain the same electron count. ***d***, Pentalayer. ***e***, van der Waals stacking of trilayers.

has a BD of 6/18 = 1/3. In principle, a stack of bilayer forming a graphite-like solid (borophite) held together by van der Waals forces can be envisaged. In the trilayer formed from the zero-HD borophene sheet (Figure 3 *b*), the middle layer acquires a BD of 6/18 by forming covalent 2c–2e bonds with both the upper and lower layers. Just as the variety possible for arrangement of holes in the 1/9 HD borophene, the position of interlayer bonds in the trilayer has many possibilities. For example, the middle layer can have all the six bonds in the repeating unit of 18 atoms with the bottom layer and in the next repeating unit all the six bonds with the top layer. Independent of the positioning of the interlayer bonds, the upper and lower layers in the trilayer are electron-rich due to the formation of only three bonds (instead of six) with the middle layer. The extra three electrons in the top and bottom layers can be dealt with by removing one boron per 18 boron atoms of the layer. This leaves the outermost layers with a HD of 1/18 and BD of 3/17 (3/18 becomes 3/17 as there is one boron less now), fulfiling a graphitic electron count. The 1/18 HD in the outer layers adds yet another variable in the positioning of the interlayer bonds. The trilayer with this combination of HDs and BDs is ready, in principle, to form another borophite stack held by van der Waals forces (Figure 3 *e*). In the same way, the middle layers in tetralayer and pentalayer borophenes with zero HD (Figure 3 *c* and *d*) can achieve the magic BD 1/3, with the peripheral (top and bottom) layers remaining electron-rich. An infinite system of this kind is possible where each layer will be bonded to its neighbouring layers above and below maintaining the same BD. The excess electrons in the two peripheral layers may either lead to different properties on the surface such as chemical reactivity or reorganize among themselves to get 1/18 HD. A similar exercise can be done starting with borophenes of different HDs or a mixture of them. Given a HD it is possible to obtain a generating formula to calculate BD. The difference in the binding energy between layers with small changes in HD will be extremely small. Interlayer stackings between monolayers of different HDs can tune the electronic properties of the bulk, aided further by the variations in BD of individual stacking depending on the electronic requirement of each monolayer. On the other hand, van der Waals stacking is possible with any of the multiple-layer structures (e.g. Figure 3 *a–d*), or any combinations of them, forming a van der Waals stacked borophite. The possibilities are endless. Obviously, the potential of borophenes for polymorphism provides an opportunity to get an effect of *n* or *p* doping in borophene just by changing the number of holes or interlayer bonds.

Comparison of experimental and computational results

The first experimental reports appeared in 2015 and 2016, where boron atoms were deposited on a silver surface

generating monoatomic layers[3,4]. While there are some variations in the results depending on the many parameters involved in the experiments, the general picture of metal template-stabilized monolayers with different HDs has emerged. The metal, boron substrate, temperature of the deposition and its rate are the variables being examined[4,5,7–10,12–15,36]. There is vigorous experimental activity in this area of borophenes, which are found to be superior in many properties than graphene. The possibility of a stable bilayer borophene has also attracted experimental attention. The first experimental observation of a bilayer on a Ag(111) surface has been published recently[23]. While the stand-alone bilayer with 1/3 BD and zero-HD is ideal, the bilayer generated by controlled atom deposition on metal surface has non-zero HD. This is anticipated; metal stabilizes the bilayer by donating electrons. Such donation is possible only if the bilayer is electron-deficient. The analysis of the data has led to the formulation of 1/9 HD with 1/4 BD in the bilayer. An interplay of the ability of the metal to donate electrons, the number of interlayer bonds (BD) and the vacancies (HD) decides the final outcome. While we discussed only 2c–2e bond between layers in forming multilayer sheets, it is possible to envisage multicentred bonds between layers, which adds further complexity. After this article was submitted to the editorial office, a second experimental report on the generation of a bilayer borophene with non-zero HD on a copper surface has appeared[37]. Attempts are on to obtain stand-alone borophene layers from metal templates[38]. Detailed theoretical and computational studies have begun to appear[35,39].

Conclusion

In this qualitative exploration of the possibilities of borophites, an analogy between boron and carbon is used to build 2D borophene sheets and stacks as graphite equivalents. Following the similarities between graphite and MgB_2 in terms of structure and electron count, a graphene-equivalent 2D boron sheet, borophene, can be constructed by replacing magnesium by boron. However, such a sheet is electron-excess and requires hexagonal holes for graphitic electron count. The HD for a monolayer borophene to be stable is 1/9, as the three extra electrons available in comparison to graphene can be reduced by removing one of the nine boron atoms. A borophene layer without any hole can form a dimeric layer with three inter layer 2c–2e bonds for every nine atoms to accommodate the graphene-like electron count. As the number of layers increases, the combinations of holes and interlayer bonds that satisfy the electron count also increases. Inter-layer bond formation can be elucidated by defining BD, which can be related to HD in a planar triangular unit. In an infinite stack, the electron requirements of the inner layers can be easily calculated. In principle, van der Waals stacking of electron-sufficient monolayers or multilayers leads to different families of borophites. Detailed theoretical as well as experimental studies are the need of the hour to explore these infinite possibility of borophenes and borophites, the latest allotropes of boron.

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