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EDITORIAL

Boron-containing compounds as labels, drugs, and theranostic agents for diabetes and its complications

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Abstract

Diabetes is a disease with a high global burden. Current strategies have failed to limit the advancement and impact of the disease. Successful early diagnosis and treatment will require the development of new agents. In this sense, boron-containing compounds have been reported as agents with the ability to reduce glycemia and lipidemia. They have also been used for labeling and measuring carbohydrates and other molecules linked to the initial stages of diabetes and its progression. In addition, certain boron compounds bind to molecules related to diabetes development and their biological activity in the regulation of elevated glycemia. Finally, it should be noted that some boron compounds appear to exert beneficial effects on diabetes complications such as accelerating wound healing while ameliorating pain in diabetic patients.

Key Words: Boron; Carbohydrates; Metabolism; Diabetes; Boronodipyrromethene

Core Tip: Diabetes is a high-global burden malady. Boron-containing compounds, from diet or administered as drugs, are promising agents to prevent or reduce progression in diabetic patients. Emerging experimental data offer promise of potential applications in the diagnosis and treatment of diabetes and its complications.

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INTRODUCTION

Diabetes is among the diseases with the highest global burden. It is the leading cause of death and disability worldwide [1]. Commissions and research groups around the world have addressed global diabetes inequity to establish concerted approaches for addressing the problem within a larger social context. This includes the development and application of new prevention approaches, early diagnosis, treatment, and minimization of complications. Efforts are currently being focused on providing a sustainable and equitable opportunity for everyone to access a healthy diet to tackle the global diabetes crisis[1,2]. Additional approaches are focused on preventing and limiting the progression of the disease in its early stages[3]. Unfortunately, the cost relating to decreasing diabetes in the global population continues to increase, despite current efforts centered on diet and exercise as prevention, because of the expense of drugs used for weight control and to regain normal glycemia levels[4-6].

Boron-containing compounds (BCC) are emerging as effective drugs in several fields of medicine[7]. In humans, boric acid and borates have been used for medicinal purposes for centuries. The applications of these and other naturally occurring BCC have expanded. Beneficial effects on hormonal, metabolism, and inflammatory systems have recently been reported[8]. BCC have been successfully incorporated into diet supplements in a growing number of countries[7].

In addition, in recent decades, five new BCC have been approved for specific medical applications with certain advantages compared to similar boron-free compounds previously utilized. Thus, bortezomib, tavaborole, crisaborole, ixazomib, and vaborbactam are now FDA-approved and used to treat certain types of cancer, mycosis, inflammatory skin diseases, and some types of urinary infections. Approximately ten other BCC are in clinical trials[9].

Hence, it is clear that medical applications of BCC are expanding in the prevention, diagnosis, and therapy of metabolic disturbances[7,10].

BCC AS LABELS OF CARBOHYDRATES FOR GLUCOSE MEASURING AND MONITORING

Some BCC react with glucose; this could be useful in the prevention, diagnosis, and control of diabetic patients. Control of glycemia limits the long-term consequences due to the disrupted glycation of vital protein structures in the heart, eyes, kidneys, nerves, and other organs. Arylboronic acids are candidates for application in the determination of glucose concentrations because of their fast and reversible formation of esters with 1,2-*cis*-diols or 1,3-diols of the glucose molecule[11].

The reactivity of boronic acids with diols has been demonstrated in multiple assays (Figure 1), including those mimicking human physiological conditions[12,13]. Boronic acids readily react with carbohydrates containing adjacent *cis* (same side)-diols that can achieve a planar arrangement which facilitates boronate ring formation as illustrated for the bolded OH groups in the α-D-glucose, and β-D-fructose furanose isomers illustrated in Figure 1[7,14,15].

Moreover, some BCC have been shown to selectivity form adducts with specific sugars. An example is described by Ramsay *et al*[16] where a boronic acid attached to a protein selectively formed adducts with α-glucose and β-fructose in the furanose forms which possess cis-diols (Figure 1). While other groups have described fluorescent boronodipyrromethene (BODIPY) conjugates with sugars[17,18].

However, BCC have failed in applications for developing non-invasive methods to measure glycemia because more efficient strategies and compounds have been developed. Despite that, multiple reports support the development of methods with high accuracy for determining glycemia *in vitro*[19,20], which is the usual procedure in the clinical monitoring of patients for the diagnosis of diabetes and its control by pharmacotherapy. A series of synthetic molecules have been developed to create a glucose sensor that would help to trigger the insulin-releasing systems; some of these are designed from an enzyme-glucose oxidase that oxidizes glucose liberating hydrogen peroxide, which then catalyzes irreversible oxidation of boronates to alcohols or phenols. However, this reaction needs to be reversible to be effective. Some arylboronic acids show a reversible binding to glucose. A boron-based, glucose sensing system might be more sensitive to changes in glucose serum levels and, thus, the release of insulin would be more accurate. Researchers are working on boronate-based materials for insulin delivery to create a more accurate artificial pancreas system[21].

There are BODIPY complexes that have emissions in different colors depending on the chemical structure of both the sugars and BODIPY[22]. It would be interesting to develop sugar-specific systems. Also, specificity among carbohydrates has been improved, including the specific forms of carbohydrates as their pyranose or furanose forms[23]. In addition, there are studies of BODIPY-sugar dyes measuring their ability to enter the cell or measuring a specific sugar in just the

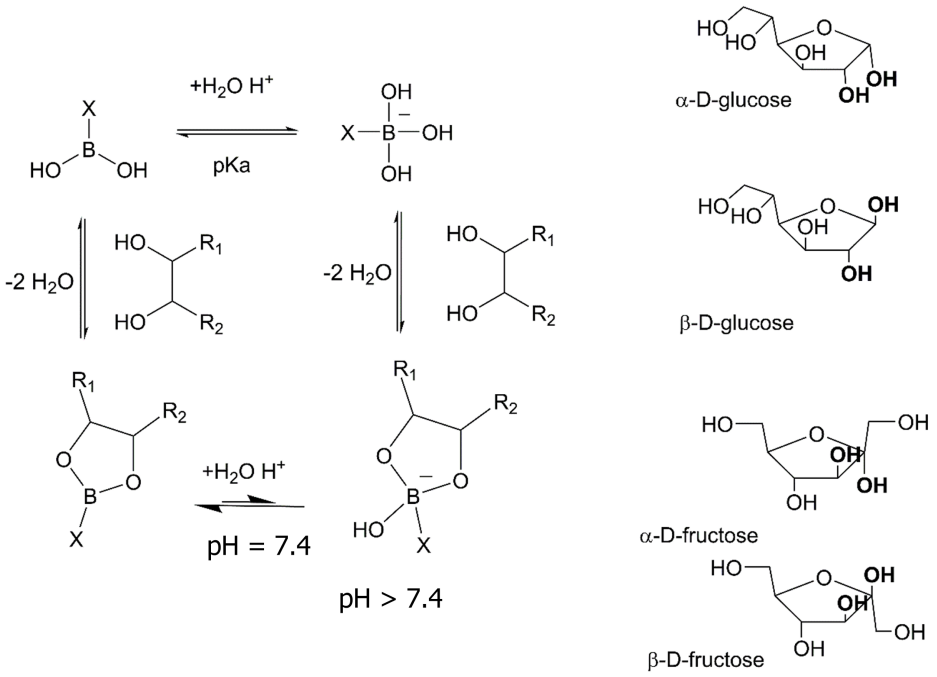

Figure 1 Interactions of boronic acids with sugars. On the left, the reaction of boronic acids with diols in aqueous solution is illustrated. The potential stability of complexes with tricoordinate boron under physiological conditions (pH = 7.4) should be noted. On the right, the structures of α and β glucose and fructose in their furanose forms.

vascular compartment[24].

BCC AS MARKERS, OR LABELING MOLECULES RELATED TO THE ORIGIN AND EVOLUTION OF DIABETES

Although there is not a clear role of BCC in human physiology, there are research protocols in which the measurement of boron is used as a potential tool for the assessment and prediction of diabetes evolution in patients. In a cross-sectional observational study, 74 patients between 25 and 75 years old with type 2 diabetes mellitus were divided into two groups: good metabolic control and poor metabolic control. They were also classified by the presence of chronic complications of diabetes mellitus. Saliva and plasma samples were obtained for mass spectrometric analysis of various trace elements, including boron. The results indicated that boron was lower in the saliva and plasma of the group with poor metabolic control. This shows that boron levels in saliva and plasma might be used to assess the control of glucose and predict the possible complications in diabetic patients; the analysis of saliva permits a less invasive and simple method to obtain results identical to the ones observed in blood analyses[25]. Other results show the correlation between the use of metformin with elevated boron levels and low activity of myeloperoxidase in the brain. It supports the importance of boron in the homeostasis of the body and its relation to the prevention of oxidative stress caused by some pathological conditions such as diabetes mellitus. Thus, brain boron levels have been postulated as a possible indicator of reduced toxic and oxidative stress[26].

Determination of boron and other endogenous molecules could improve metabolic dysfunction in diabetic patients. As an example, it should be mentioned that some researchers have found that boron and vitamin D are important in the secretion of insulin and the regulation of glycemia. In this sense, studies have demonstrated that, after administration of dietary boron, the vitamin D3 serum concentration increases; boron was linked to an enhanced response for limiting hyperglycemia[8,10].

BCC also can act as labeling molecules for other processes well-known in the origin and evolution of diabetes; this approach could be useful for the diagnosis and monitoring of diabetic patients.

Currently, the glycated hemoglobin (HbA1c) measurement is considered important in the diagnosis of diabetes and for the patient follow-up, based on the capacity of the heme group in the hemoglobin to bind to the glucose present in the plasma. This glucose is carried on the red blood cells, which have an average life cycle of about 2-3 months, and the value does not change with the daily fluctuations in the glucose serum levels. Studies similar to the closed-loop glucose sensor were carried out for HbA1c. The selectivity between a sensing system based on IgG and a new system using a 4-vinylphenylboronic acid (VPBA) coated nanofilm (using concentrations of 30 mg/mL, 50 mg/mL, and 120 mg/mL HbA1c) was carried out in artificial plasma. It was observed that the VPBA nanofilm had a selectivity coefficient 7.30 times more selective than the IgG-based system, 17.33 times more than the human serum albumin, and 34.66 times more

Table 1 Boron-containing compounds exert effects similar to those described for drugs currently used as therapy in diabetes patients

Type of drug	Example in use	BCC acting in a similar or related way	Ref.
Biguanides	Metformin	Boron-containing biguanides have been synthesized and some have shown biological effects. Some BCC is being used in biomaterials for metformin release	Anderson *et al*[59], 1995; Ghosh *et al*[60], 1998; Lai *et al*[61], 2023
GLP-1 receptor agonist	Exenatide	No BCC has been designed with a structural analogy to known GLP-1 agonists. However, some BCC seems to be able to modify the incretins serum levels	Das *et al*[9], 2022; Ri *et al*[7], 2023
DPP-IV inhibitors	Sitagliptin	Dutogliptin, talabostat, PC06R58, and PC06R108 are potent uncompetitive DPP-IV inhibitors	Wu *et al*[55], 2021; Prajapati *et al*[62], 2024
SGLT2 inhibitors	Dapagliflozin	DX-B-DA, a fluorophore-dapagliflozin dyad has been tested as theranostic agent. It can bind the SGLT2	Yu *et al*[63], 2021
Sulfonylureas	Glibenclamide	Glibenclamide-BCC acts as a high-affinity blocker of pancreatic β-cell K_{ATP} currents. Non-specific binding limited its use	Zünkler *et al*[64], 2004
α-glucosidase inhibitor	Acarbose	Some azaborininones exhibit moderate to good inhibitory effects against glucosidase to acarbose used as a reference standard	Mphahlele *et al*[65], 2021
Insulin	Insulin	Several BCC result in changes in insulin serum concentrations. Boric acid and phenyl-boronic acids diminish the increase of insulin release (and the total weight of visceral fat) in rats exposed to a high-fat diet and streptozotocin. The insulin release could be controlled by complexes with boronic acids	López-Cabrera *et al*[42], 2018; Banach *et al*[21], 2021; Kikuchi *et al*[66], 2021

GLP-1: Glucagon-like peptide; CPP-IV: Dipeptidyl peptidase-4; SGLT2: Sodium-glucose cotransporter 2; BCC: Boron-containing compound.

than hemoglobin[27].

Reactions of BODIPY with some membranes as well as intracellular and plasma lipids have been reported as biomarkers of use in diabetes[28-31]. Furthermore, a mix of BODIPYs could be used to determine more than one metabolite linked to diabetes evolution. For example, pyridine-extended BODIPY and BODIPY-cholesterol have been shown to provide reliable and accurate measurements of glucose and cholesterol from plasma samples[20,32].

Other BODIPY dyes have been described for measuring insulin in monomers and oligomers[33], carbohydrate complexes[34], glucagon, and its receptor[35,36]. Also, the design of dyes offers potential methods for using BODIPY targeting to detect the expression and function of glucose transporters among these are SGLT2[37], and GLUT4[38].

BCC ACT ON THE GLUCOSE AND LIPIDS SERUM LEVELS

The effects of BCC on metabolic parameters in mammals have been recently reviewed[7,8]. Some differences have been observed depending on the administered form of the BCC[39]. It has also been reported that BCC intake exerts ameliorative effects on the metabolism disruption induced by streptozotocin[40-42].

Demirdogen *et al*[43] observed lower serum boron levels in diabetic patients when compared to healthy patients. They found a negative correlation between serum levels of HbA1c and serum boron levels in normal *vs* obese diabetic patients. Kuru *et al*[44] reported that a boron-rich diet induced a decrease in LDL, VLDL cholesterol, triglyceride levels, body weight, body fat, and body mass index. BBC might regulate lipid profile secondary to immunomodulatory effects, as has been suggested for calcium fructoborate[45-47].

In vitro, assays with human adipose-derived stem cells showed that BCC can restrain the expression of adipogenesis-related genes and proteins, like the peroxisome proliferator activated receptor γ. Apparently by regulating decisive growth factors such as β-catenin[48]. Moreover, high concentrations of sodium borate (68 μM to 340 μM) resulted in a progressive decrease in lipid deposition[49,50].

Currently, there are several drugs available for diabetes treatment using modulation of glycemia; and several BCC have shown an ability to act on the same targets (Table 1, Figure 2). Examples are those with action on specific enzymes related to diabetes. Regarding effects on carbohydrate metabolism, dutogliptin is a dipeptidyl peptidase IV inhibitor (the enzyme responsible for the degradation of incretins)[51,52], which is safely used in humans[51,53]. Another BCC, such as talabostat, also acts on dipeptidyl peptidases and other proteins and is an attractive metabolism regulator[54,55]. Some BCC act on glucose transporters, in a similar way to gliflozin. Cakir *et al*[41] suggested BA could affect glucose transporters. Recently, some BCC have shown an ability to bind the SGLT2[56,57].

BCC have the potential to act on emerging identified targets, such as AN2898 acting as a PDE4 inhibitor since it could be applied to metabolic modulation in addition to its well-known action on smooth muscle[58] (Table 1[59-66]).

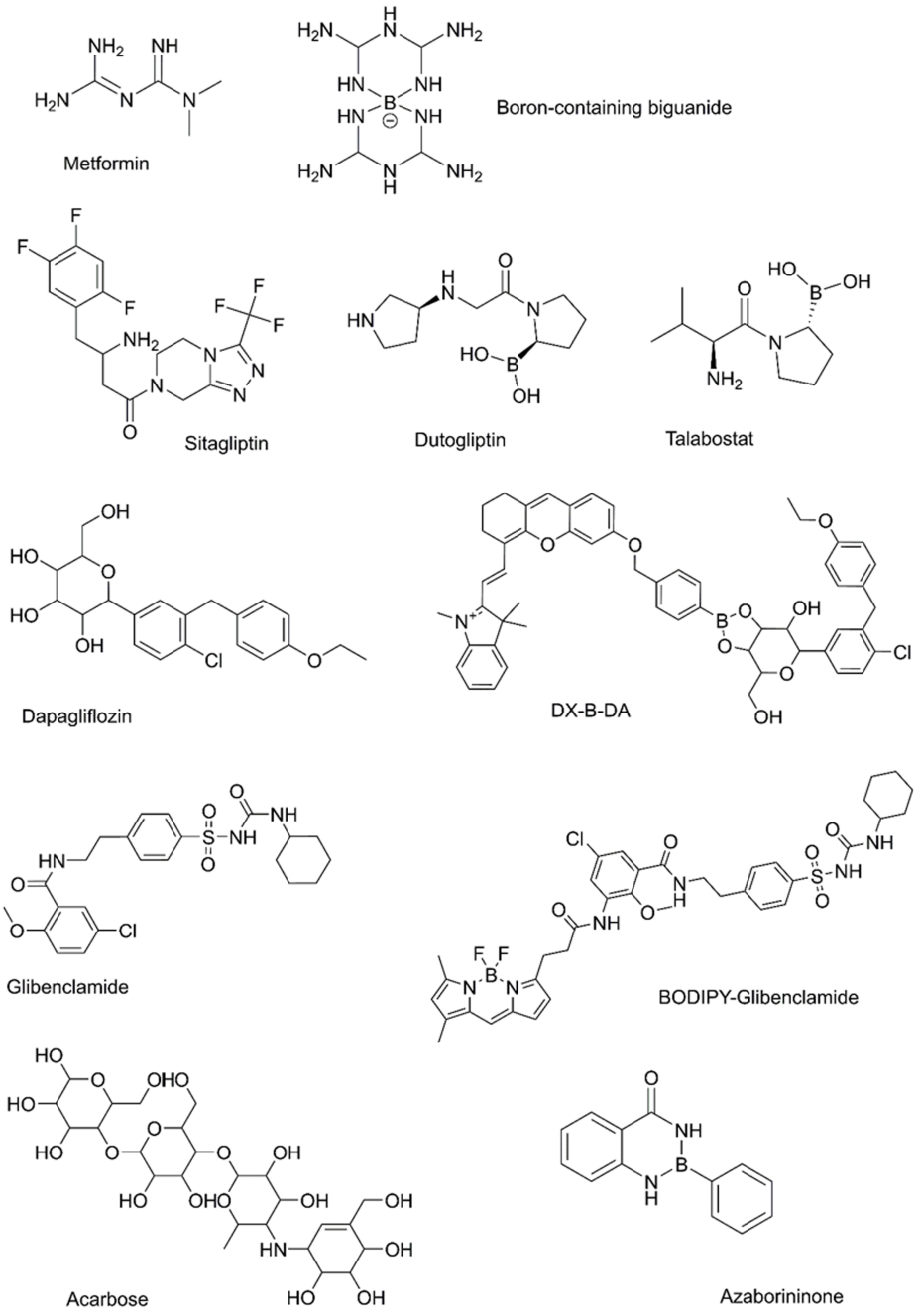

Figure 2 Examples of boron-containing compounds (on the right) whose activity parallels that of current drugs (on the left) for diabetes treatment. BODIPY: Boronodipyrromethene.

BCC AS POTENTIAL AGENTS IN DIABETES COMPLICATIONS

There are a wide variety of diabetes complications such as those related to cardiovascular dysfunction (including amputations and infective diseases), renal failure, retinopathy, and neuropathy[67-69].

Some BCC are effective in the prevention of cardiovascular diseases, mainly modulating chronic inflammation and oxidative stress; there is evidence that BCC modulate cardiac and vascular remodeling[70-73]. Moreover, dietary boron appears to be associated with a healthier diet and seems to be related to lower BMI and a more favorable cardio-metabolic risk profile[74].

Specific BCC are attractive as agents to treat diabetic complications (Figure 3). Significantly, there is evidence regarding their use in the treatment of the diabetic foot. In this sense, it has been observed that the boric acid in a 3% formulation has a great effect on the wound healing process in humans[75,76]; it is associated with important healing mechanisms, such as cell migration, collagen deposition, and superoxide dismutase activity. A study evaluated the effect of boric acid and sodium tetraborate in dermal cell cultures, showing that both compounds, at intermediate concentrations, increased the proliferation and migration of dermal cells[77]. In rats induced with diabetes-like syndromes, BCC (boric acid, sodium tetraborate, phenylboronic-based gel, or hexagonal boron nitrides) improved or accelerated wound healing[78-80]. In addition, a recent research protocol studied the wound-healing effect of BBC; 171 participants with foot ulcers

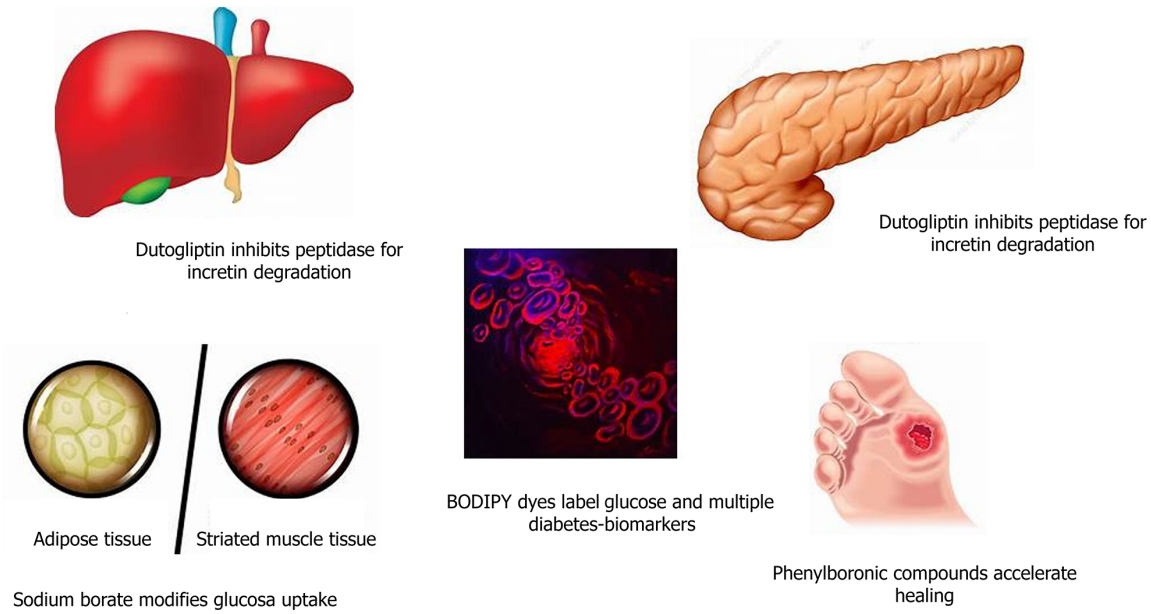

Figure 3 Examples of boron-containing compound applied to treat diabetes and its complications. BODIPY: Boronodipyrromethene.

were divided into two groups, one being the control group and the other using a formulated gel with 3% sodium pentaborate twice a day. The treatment, using topical administration, helped to decrease ulceration. Fewer treatments were necessary for the intervention group than the control group and recurrence was not observed in the intervention group; it was suggested that the acidic boron environment promotes angiogenesis and epithelization in the ulceration and increases the antimicrobial activity[81]. The attractiveness of BCC in diabetic foot ulcer treatment goes beyond wound healing acceleration because BCC have also been shown to modulate pain and exert antimicrobial effects[82,83].

Regarding nephropathy, no clear studies exploring BCC in the kidneys of diabetic patients have been reported. However it is well-known that these compounds exert effects in nephron remodeling[84], probably by acting in some enzymes (such as carbonic anhydrase)[85,86]; additionally, they have been used to treat commonly associated maladies, such as lithiasis[87].

No studies have been reported evaluating the retinopathy or neuropathy observed in diabetic patients but the effect of multiple BCC as neuroprotective agents has been described, including the limitation of oxidative or inflammatory damage or maintaining the production of local or systemic neurotrophic agents[88,89]. This is relevant due to the increased interest in neurological disorders and cognitive or motor deficits linked to metabolic disturbance[89].

It should be kept in mind that diabetes not only affects the glucose serum levels, it also predisposes tissues to additional abnormalities. Osteoporosis is a bone health problem associated with diabetes mellitus due to absorption loss and increased secretion of minerals such as calcium, magnesium, phosphorus, and vitamin D3; all of which are required for normal development of bone structure. It has been demonstrated that BCC improve the utilization of these minerals and vitamins in diabetic mice. A proposed mechanism of the action of boron against osteoporosis is the hydroxylation and protection of the steroid hormones from fast degradation while influencing the synthesis of vitamin D[10].

BCC AS POTENTIAL THERANOSTIC AGENTS

Some compounds are attractive because they can bind molecules that are linked to the onset and evolution of diabetes. However unexpected and unintended biological actions have been reported in the regulation of the metabolism of a cellular system or an organism. Conceptually, the theranostic approach should include simultaneous exploration of the agent as a drug and its diagnostic effect, but also the consideration (by authorities) of the coupled combination in the drug approval process[90]. In fact, to the best of our knowledge, no BCCs are registered to date as theranostic agents for diabetes (some theranostic-BCC have been reported for cancer treatment[91,92]) and some of the recent reports demonstrate the potential role of some BCC in this field.

Examples of BCC with potential as theranostic agents include BCC-labeled enzymes involved in carbohydrate metabolism. Thus, there are chromophoric BODIPY/glycoside systems for use within a specific cellular compartment, designed by chemically modifying the BODIPY skeleton to use them for *in vivo* imaging with luminescence being shifted to therapeutic purposes[93]. Likewise, BCC labeling of membrane carbohydrate transporters (modifying the transportation rate). These include dyad boronated complexes acting as precursors of near-infrared emitting molecules and prodrug-modulating carbohydrate transport through the membrane which are promising agents in early kidney dysfunction detection[63]. Also, certain BCC reach beta-cells in pancreatic islets and can act as biomarkers and modify the release; thus boronophenylalanine derivatives are efficiently captured in pancreatic cells[94], while structurally related BCC could modulate insulin release[66].

CONCLUSION

Several BCC affect aspects of metabolism in humans. Moreover, BCC seem to have potential applications for improving the performance of available molecules in the prevention, diagnosis, and treatment of diabetes. This is confirmed by the reactivity of BCC on *cis*-diols of sugars and other chemical moieties related to the origin and evolution of diabetes and its complications. In this sense, notable effects of some BCC on the concentration of carbohydrates in plasma, the vascular system, inflammation, and wound healing are particularly attractive for studies in and application to diabetic patients.

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FOOTNOTES

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