

Reactions of Substituted Furan-2-carboxaldehydes and Furo[*b*]pyrrole Type Aldehydes with Benzothiazolium Salts

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Abstract: A series of new *push-pull* compounds were synthesised by reaction of 5-aryl-furan-2-carboxaldehydes and furo[*b*]pyrrole type aldehydes with benzothiazolium salts. These new condensation products represent highly conjugated systems that have potential biological activity. The reaction of furo[*b*]pyrrole type aldehydes with benzothiazolium salts give potential precursors of cyanine dyes.

Keywords: 5-Arylfuran-2-carboxaldehydes; 2-formylfuro[3,2-*b*]pyrrole-5-carboxylates; 2-formylfuro[2,3-*b*]pyrrole-5-carboxylate; benzothiazolium salts, ¹H- and ¹³C-NMR spectra.

Introduction

Many compounds containing the benzothiazole moiety are reported to be biologically active and good structure-activity relations have been found using QSAR studies [1,2]. High antimicrobial activity of benzothiazolium salts has been reported for systems bearing electron-donating substituents at C2 and the activity increases if the substituent is bound by a conjugated polyene bridge [3]. The *push-pull* system is responsible for the high biological activity observed in these compounds. It has also been reported that these compounds can also be used as precursors of cyanine dyes [4].

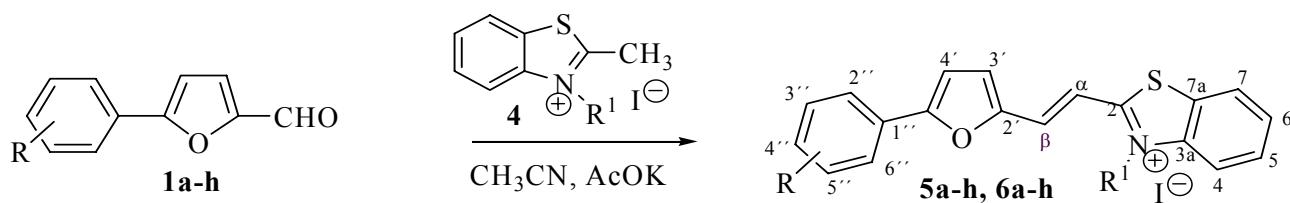
In the past we have been interested in the synthesis and reactions of 5-arylfuran-2-carboxaldehydes **1** [5]. Many of the published condensation products of 5-arylfuran-2-carboxaldehydes are also reported to be biologically active compounds [6,7] or can be used as intermediates in organic synthesis [8,9]. Furo[*b*]pyrrole type aldehydes **2** and **3** are heteroanalogues of the pentalene dianion [10]. Their full synthesis and properties were described recently [11]. Only a few condensation products of furo[*b*]pyrrole type aldehydes have been described [12, 13]. We have recently published in this journal the results of the study of the reactions of substituted furan-2-carboxaldehydes and furo[*b*]pyrrole type aldehydes with hippuric acid [14].

To the best of our knowledge 5-aryl furan-2-carboxaldehydes **1a-h**, furo[3,2-*b*]pyrrole type aldehydes **2** and furo[2,3-*b*]pyrrole type aldehyde **3** have never been used in reactions with benzothiazolium salts. This fact has prompted us to synthesise a series of new condensation products of 5-aryl furan-2-carboxaldehydes **1a-h** with 2,3-dimethyl-benzothiazolium iodide (**4a**), 3-ethyl-2-methylbenzothiazolium iodide (**4b**) and 3-benzyl-2-methylbenzothiazolium bromide (**4c**) under Knoevenagel reaction conditions. In this way we have obtained new highly conjugated systems **5a-7h** containing furan and benzothiazole rings with potential biological activity. Further, we also describe condensation products of methyl 2-formyl-4*H*-furo[3,2-*b*]pyrrole-5-carboxylate (**2a**) with 2,3-dimethylbenzothiazolium iodide (**4a**) and 3-benzyl-2-methylbenzothiazolium bromide (**4c**). The 4-R derivatives **2b-d** and methyl 2-formyl-6-methoxymethylfuro[2,3-*b*]pyrrole-5-carboxylate (**3**) were reacted with 3-benzyl-2-methylbenzothiazolium bromide (**4c**) to obtain a series of new condensation products **8a-d and 9**. We have thus prepared 28 new compounds with predictable biological activity containing benzothiazole and substituted furan moieties. The structures of the products are fully supported by spectral data.

Results and Discussion

The reactions of 5-aryl furan-2-carboxaldehydes **1a-h** with benzothiazolium salts **4** were carried out under various conditions. In the first case (Method A) acetonitrile was used as solvent and potassium acetate as support/catalyst in the reaction of 5-aryl furan-2-carboxaldehydes **1a-h** with benzothiazolium salts **4a,b**, as shown in Scheme 1 [19].

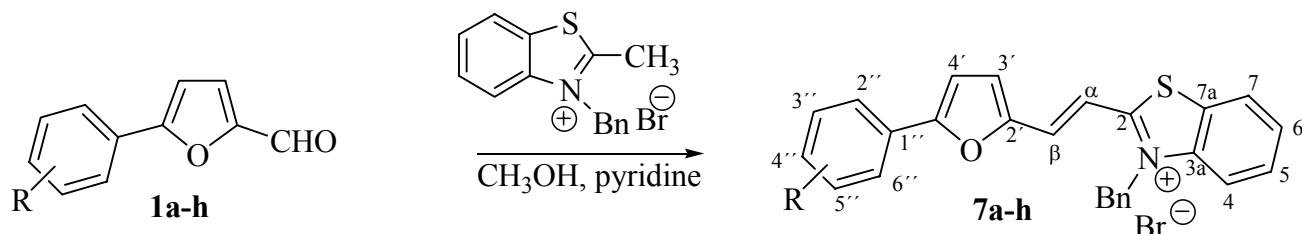
Scheme 1



In formulas **1**, **5**, **6** R for **a**: 4-Cl, **b**: 4-Br, **c**: 4-NO₂, **d**: 3-NO₂, **e**: 2-NO₂, **f**: 3-CF₃, **g**: 2-Br, **h**: 2,4-Cl₂,
In formulas **4**: R¹ = CH₃ or C₂H₅; **5**: R¹ = CH₃; **6**: R¹ = C₂H₅

The products **5a-6h** were obtained in 37-88% yield. The yield of the final product was dependent on the benzene ring substitution of the starting aldehydes **1a-h**. The lowest yields were obtained when the reaction was carried out with **1e** and **1g**, which possess *ortho*-substituents on the benzene ring. The highest yield was obtained with compound **1c** having a 4-nitrophenyl group at the C5 position of the furan ring. A disadvantage of this procedure was the low solubility of the starting compounds in the acetonitrile which leads to low yields of the condensation products and in some cases the unreacted starting material was isolated along with the products. The condensation products had to be separated from the crude reaction mixtures by crystallisation and recrystallised to remove the starting material and thus give analytically pure compounds. Due to this experimental observation, for the subsequent reactions of compounds **1a-h** with 3-benzyl-2-methylbenzothiazolium bromide (**4c**) we changed the reaction conditions.

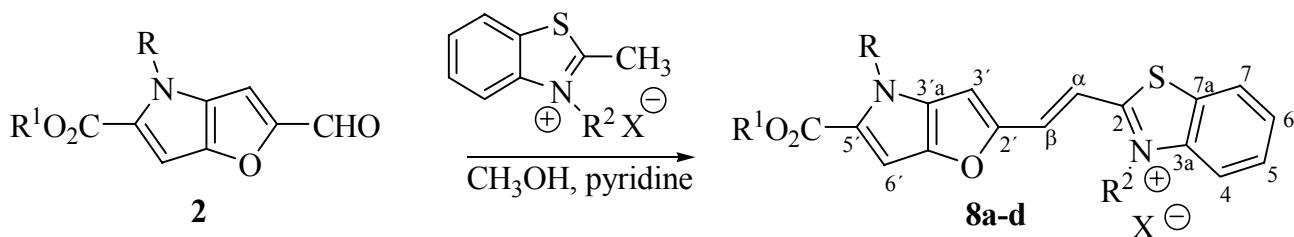
Scheme 2



In formulas **1**, **7** R for **a**: 4-Cl, **b**: 4-Br, **c**: 4-NO₂, **d**: 3-NO₂, **e**: 2-NO₂, **f**: 3-CF₃, **g**: 2-Br, **h**: 2,4-Cl₂,

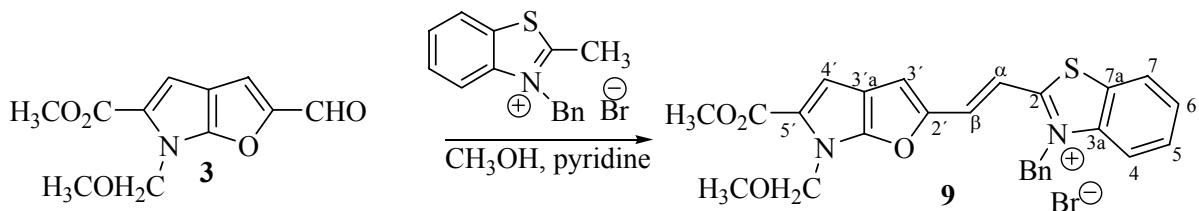
The published conditions using methanol as solvent and pyridine as catalyst (Method B) for the reactions shown in Scheme 2 was used [1,2]. With this procedure, products **7a-h** were obtained as analytically pure compounds directly from the reaction mixtures in higher yields (43-91%). The same situation as in the reaction with acetonitrile was observed. *Ortho*-substituted benzene rings in the starting 5-aryl furan-2-carboxaldehydes **1e**, **1g**, **1h** gave the lowest yields of products (~50%) and the 5-aryl furan-2-carboxaldehydes **1a-c** with *para*-substituted benzene rings gave the highest yields of products (~80%). The condensations of furo[*b*]pyrrole type aldehydes **2** and **3** with benzothiazolium salts **4a,c** lead to the corresponding products **8a-d** and **9** in satisfactory yields (~40-60%) (Schemes 3, 4). The reactions were carried out in refluxing methanol with pyridine as catalyst. The character of furo[*b*]pyrrole type aldehydes [11] causes lower reactivity of carbonyl group in the C2 position in condensation reaction than is the case for 5-aryl furan-2-carboxaldehydes where the carbonyl group at C2 of the furan ring is activated by a benzene ring substituent in the C5 position.

Scheme 3



8	R	R¹	R²	X
a	H	CH ₃	CH ₃	I
b	H	CH ₃	Bn	Br
c	CH ₃	C ₂ H ₅	Bn	Br
d	Bn	C ₂ H ₅	Bn	Br

Scheme 4



All the condensation products are high melting stable solids that are sparingly soluble in common solvents. They are characterised by their colour. The absorption maximum appears in the VIS spectrum around 460 - 470 nm **5a-7h** and 480 - 500 nm **8a-d** and **9**.

The ¹H- and ¹³C-NMR spectral data of all synthesised products are collated in the Experimental section. The ¹H-NMR spectra of the synthesised compounds display doublets for double bond protons in the 7.50-7.70 ppm region for H- α and 8.10-8.20 ppm for H- β , respectively. The coupling constants of the H- α and H- β protons are 15.2-15.5 Hz, which proves the *E* configurations of the condensation products. In the ¹³C-NMR spectra the chemical shifts for the C- α double bond carbons are observed at 110-111 ppm and C- β at 133-135 ppm. The chemical shifts of protons were assigned using gs-H,H-COSY (gradient selected Correlated Spectroscopy) measurements which provided the proton-proton connectivity. ¹³C-NMR chemical shift assignments were straightforward using gs-HSQC (gradient selected Heteronuclear Quantum Coherence) and gs-HMBC (gradient selected Heteronuclear Multiple Bond Correlation).

Conclusions

The syntheses of 3-methyl-2-[(*E*)-2-(5-aryl furan-2-yl)vinyl]-1,3-benzothiazolium iodides (**5a-h**), 3-ethyl-2-[(*E*)-2-(5-aryl furan-2-yl)vinyl]-1,3-benzothiazolium iodides (**6a-h**), 3-benzyl-2-{(*E*)-2-(5-aryl furan-2-yl)vinyl}-1,3-benzothiazolium bromides (**7a-h**), 2-{(*E*)-2-[5-(methoxycarbonyl)-4*H*-furo[3,2-*b*]pyrrol-2-yl]vinyl}-3-methyl-1,3-benzothiazolium iodide (**8a**), 3-benzyl-2-{(*E*)-2-[5-(ethoxy / methoxycarbonyl)- 4*H*-, 4-methyl and 4-benzyl furo[3,2-*b*]pyrrol-2-yl]vinyl}-1,3-benzothiazolium bromides (**8b-d**) and 3-benzyl-2-{(*E*)-2-[5-(methoxycarbonyl)-6-methoxymethylfuro[2,3-*b*]pyrrol-2-yl]vinyl}-1,3-benzothiazolium bromide (**9**) are described. All synthesised compounds were characterised by spectroscopic measurements and were shown to be the pure *E* isomers.

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Experimental

General

The compounds **1a-h** were prepared according to references [15, 16]. Furo[3,2-*b*]pyrrole type aldehydes **2** and **3** were prepared according to the literature [12, 17, 18]. Benzothiazolium salts **4a-c** were prepared according to reference [19]. Melting points were determined using a Kofler hotplate apparatus and are uncorrected. All solvents were pre-distilled and dried appropriately prior to use. Concentration and evaporation refer to the removal of volatile materials under reduced pressure using a Büchi Rotovapor. Substances stated to be identical were so with respect to m.p.s, mixed m.p.s and IR spectra. Elemental analyses were determined using a Carlo Erba CHNS-OEA 1108-Elemental Analyser. UV spectra were measured on WPA UV/VIS Diode-Array spectrophotometer (Cambridge, UK) in methanol (λ_{max} (log ε); λ_{max} in nm, ε in $\text{m}^2\text{mol}^{-1}$). ^1H and ^{13}C NMR spectra were obtained using for: **5a-h**, **6a-h** BRUKER B-ACS (400 and 100 MHz), **5e** BRUKER AMX (360 and 90 MHz), **7d-h** VARIAN VRX (300 MHz, 75 MHz), **7a-c**, **8a-d** and **9** BRUKER AVANCE (500 MHz, 125 MHz) in $\text{DMSO}-d_6$ with TMS as an internal standard reference. The methods used for the purpose of NMR assignment were gs-H,H-COSY, gs-HSQC and gs-HMBC. Coupling constants (*J*) are quoted to the nearest 0.3 Hz. Chemical shifts (δ -scale) are quoted in parts per million and the following abbreviations are used: s = singlet; d = doublet; t = triplet; q = quartet; m = multiplet; br = broad.

General procedures for synthesis of the condensation products.

Method A: A mixture of 5-aryl furan-2-carboxaldehyde **1a-h** (2 mmol), benzothiazolium salt **4a,b** (2.4 mmol) and a catalytic amount of potassium acetate (0.2 mmol) in dry acetonitrile (10-15 mL) was refluxed with stirring 1-5 h (monitored by TLC using 4:1 chloroform-methanol). After cooling down to room temperature the solid product was separated by filtration, washed with acetonitrile and methanol, dried and purified by crystallisation. Products **5a-6h** were prepared using this method.

Method B: A mixture of 5-aryl furan-2-carboxaldehyde **1a-h** (2 mmol) or furo[*b*]type aldehyde **2** or **3** (2 mmol) and benzothiazolium salt **3a,c** (2.4 mmol) in methanol (15 mL) was refluxed for 1-4 h in the presence of a catalytic amount of pyridine (15 mg). Reactions were monitored by TLC (4:1 chloroform-methanol). After cooling down to room temperature the solid product was separated by filtration, washed with methanol and dried. Products **7a-9** were prepared by this procedure.

2-{(E)-2-[5-(4-Chlorophenyl)furan-2-yl]vinyl}-3-methyl-1,3-benzothiazolium iodide (5a**).** Reaction time: 85 min; yield 57%; m.p. 249-254°C (methanol); Calc. for C₂₀H₁₅ClINOS (479.76): 50.07% C, 3.15% H, 7.39% Cl, 2.92% N, 6.68% S; found: 50.22% C, 3.12% H, 7.43% Cl, 2.89% N, 6.71% S; UV: 463 (3.32); ¹H-NMR (400 MHz): 8.42 (d, 1H, J_(4,5) = 8.1 Hz, H-4), 8.24 (d, 1H, J_(6,7) = 8.4 Hz, H-7), 8.08 (d, 2H, J_(2'',3'') = 8.5 Hz, H-2'', H-6''), 8.12 (d, 1H, J_(α,β) = 15.6 Hz, H-β), 7.87 (t, 1H, J_(4,5) = 8.1 Hz, J_(5,6) = 8.3 Hz, H-6), 7.78 (t, 1H, J_(5,6) = 8.3 Hz, J_(6,7) = 8.4 Hz, H-5), 7.74 (d, 1H, J_(α,β) = 15.6 Hz, H-α), 7.61 (d, 2H, J_(2'',3'') = 8.5 Hz, H-3'', H-5''), 7.47 (d, 1H, J_(3',4') = 3.7 Hz, H-3'), 7.43 (d, 1H, J_(3',4') = 3.7 Hz, H-4'), 4.36 (s, 3H, N⁺CH₃); ¹³C-NMR (100 MHz): 170.9 (C-2), 157.0 (C-5'), 150.5 (C-2'), 142.1 (C-3a), 134.1 (C-4''), 133.1 (C-β), 129.3 (C-6), 129.2 (C-2''), 128.2 (C-5), 127.6 (C-7a), 127.5 (C-1''), 126.7 (C-3''), 124.2 (C-7), 123.7 (C-3'), 116.7 (C-4), 111.1 (C-4'), 110.1 (C-α), 36.2 (N⁺CH₃).

2-{(E)-2-[5-(4-Bromophenyl)furan-2-yl]vinyl}-3-methyl-1,3-benzothiazolium iodide (5b**).** Reaction time: 160 min; yield 57%; m.p. 255-258°C (ethanol); Calc. for C₂₀H₁₅BrINOS (524.21): 45.82% C, 2.88% H, 15.24% Br, 2.67% N, 6.12% S; found: 46.05% C, 2.92% H, 15.19% Br, 2.62% N, 6.17% S; UV: 463 (3.69); ¹H-NMR (400 MHz): 8.42 (d, 1H, J_(4,5) = 8.0 Hz, H-4), 8.24 (d, 1H, J_(6,7) = 8.4 Hz, H-7), 8.00 (d, 2H, J_(2'',3'') = 8.4 Hz, H-2'', H-6''), 8.11 (d, 1H, J_(α,β) = 15.2 Hz, H-β), 7.87 (t, 1H, J_(5,6) = 7.8 Hz, J_(6,7) = 8.4 Hz, H-6), 7.78 (t, 1H, J_(4,5) = 8.0 Hz, J_(5,6) = 8.3 Hz, H-5), 7.73 (d, 1H, J_(α,β) = 15.2 Hz, H-α), 7.73 (d, 2H, J_(2'',3'') = 8.4 Hz, H-3'', H-5''), 7.47 (d, 1H, J_(3',4') = 3.7 Hz, H-3'), 7.43 (d, 1H, J_(3',4') = 3.7 Hz, H-4'), 4.37 (s, 3H, N⁺CH₃); ¹³C-NMR (100 MHz): 170.9 (C-2), 157.1 (C-5'), 150.5 (C-2'), 142.0 (C-3a), 132.1 (C-2''), 133.1 (C-β), 129.3 (C-6), 128.2 (C-5), 127.8 (C-4''), 127.6 (C-7a), 126.8 (C-3''), 124.2 (C-7), 122.9 (C-1''), 123.7 (C-3'), 116.6 (C-4), 111.1 (C-4'), 110.1 (C-α), 36.3 (N⁺CH₃).

3-Methyl-2-{(E)-2-[5-(4-nitrophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium iodide (5c). Reaction time: 270 min; yield 84%; m.p. 242-246°C (ethanol); Calc. for C₂₀H₁₅IN₂O₃S (490.32): 48.99% C, 3.08% H, 5.71% N, 6.54% S; found: 49.38% C, 3.14% H, 5.91% N, 6.61% S; UV: 453 (3.64); ¹H-NMR (400 MHz): 8.44 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.37 (d, 2H, $J_{(2'',3'')} = 8.0$ Hz, H-3'', H-5''), 8.30 (d, 2H, $J_{(2'',3'')} = 8.0$ Hz, H-2'', H-6''), 8.27 (d, 1H, $J_{(6,7)} = 8.7$ Hz, H-7), 8.16 (d, 1H, $J_{(\alpha,\beta)} = 15.6$ Hz, H-β), 7.89 (t, 1H, $J_{(4,5)} = 8.0$ Hz, $J_{(5,6)} = 7.7$ Hz, H-6), 7.85 (d, 1H, $J_{(\alpha,\beta)} = 15.6$ Hz, H-α), 7.80 (t, 1H, $J_{(5,6)} = 7.7$ Hz, $J_{(6,7)} = 8.7$ Hz, H-5), 7.66 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.52 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 4.37 (s, 3H, N⁺CH₃); ¹³C-NMR (100 MHz): 171.4 (C-2), 156.0 (C-5'), 152.4 (C-2'), 147.1 (C-1''), 142.7 (C-3a), 135.0 (C-4''), 133.5 (C-β), 130.1 (C-6), 129.0 (C-5), 128.5 (C-7a), 126.3 (C-3''), 125.0 (C-2''), 124.9 (C-7), 123.6 (C-3'), 117.0 (C-4), 114.5 (C-4'), 112.2 (C-α), 37.0 (N⁺CH₃).

3-Methyl-2-{(E)-2-[5-(3-nitrophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium iodide (5d). Reaction time: 120 min; yield 82%; m.p. 220-224°C (ethanol); Calc. for C₂₀H₁₅IN₂O₃S (490.32): 48.99% C, 3.08% H, 5.71% N, 6.54% S; found: 49.26% C, 3.21% H, 5.87% N, 6.65% S; UV: 449 (3.36); ¹H-NMR (400 MHz): 8.76 (s, 1H, H-2''), 8.49 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.43 (d, 1H, $J_{(5'',6'')} = 8.1$ Hz, H-6''), 8.27 (d, 1H, $J_{(6,7)} = 8.0$ Hz, H-7), 8.26 (d, 1H, $J_{(4'',5'')} = 8.2$ Hz, H-4''), 8.1 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H-β), 7.88 (t, 1H, $J_{(4,5)} = 8.0$ Hz, $J_{(5,6)} = 7.5$ Hz, H-6), 7.83 (t, 1H, $J_{(4'',5'')} = 8.2$ Hz, $J_{(5'',6'')} = 8.1$ Hz, H-5''), 7.83 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H-α), 7.79 (t, 1H, $J_{(5,6)} = 7.5$ Hz, $J_{(6,7)} = 8.0$ Hz, H-5), 7.65 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.51 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 4.38 (s, 3H, N⁺CH₃); ¹³C-NMR (100 MHz): 170.8 (C-2), 155.3 (C-5'), 151.0 (C-2'), 148.5 (C-3''), 141.9 (C-3a), 132.9 (C-β), 130.7 (C-6''), 130.6 (C-4''), 130.1 (C-1''), 129.3 (C-6), 128.2 (C-5), 127.7 (C-7a), 124.9 (C-7), 124.1 (C-5''), 122.9 (C-3'), 116.6 (C-4), 119 (C-2''), 112.3 (C-4'), 111 (C-α), 36.2 (N⁺CH₃).

3-Methyl-2-{(E)-2-[5-(2-nitrophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium iodide (5e). Reaction time: 7 hours (synthesised without using a catalyst); yield 37%; m.p. 240-243°C (ethanol); Calc. for C₂₀H₁₅IN₂O₃S (490.32): 48.99% C, 3.08% H, 5.71% N, 6.54% S; found: 48.66% C, 3.19% H, 5.82% N, 6.48% S; UV: 439 (3.49); ¹H-NMR (360 MHz): 8.43 (d, 1H, $J_{(4,5)} = 8.7$ Hz, H-4), 8.25 (d, 1H, $J_{(6,7)} = 8.6$ Hz, H-7), 8.12 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H-β), 8.1 (d, 1H, $J_{(5'',6'')} = 7.9$ Hz, H-6''), 8.02 (d, 1H, $J_{(3'',4'')} = 8.1$ Hz, H-3''), 7.85 (t, 1H, $J_{(4,5)} = 8.7$ Hz, $J_{(5,6)} = 7.7$ Hz, H-6), 7.84 (t, 1H, $J_{(4'',5'')} = 8.6$ Hz, $J_{(5'',6'')} = 7.9$ Hz, H-5''), 7.74 (t, 1H, $J_{(3'',4'')} = 8.1$ Hz, $J_{(4'',5'')} = 8.6$ Hz, H-4''), 7.70 (t, $J_{(5,6)} = 7.7$ Hz, $J_{(6,7)} = 8.6$ Hz, 1H, H-5), 7.51 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H-α), 7.48 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.22 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 4.30 (s, 3H, N⁺CH₃); ¹³C-NMR (100 MHz): 171 (C-2), 152.9 (C-5'), 151.8 (C-2'), 147.7 (C-2''), 142.4 (C-3a), 133.3 (C-β), 133.3 (C-5''), 131.3 (C-4''), 130.1 (C-6''), 129.9 (C-6), 128.9 (C-5), 128.3 (C-7a), 124.8 (C-3''), 124.7 (C-7), 122.5 (C-3'), 122 (C-1''), 117.2 (C-4), 114.3 (C-4'), 111.5 (C-α), 36.3 (N⁺CH₃).

3-Methyl-2-{(E)-2-[5-[3-(trifluoromethyl)phenyl]furan-2-yl]vinyl}-1,3-benzothiazolium iodide (5f). Reaction time: 150 min; yield 46%; m.p. 242-246°C (methanol); Calc. for C₂₁H₁₅F₃INOS (513.32): 49.14% C, 2.95% H, 2.73% N, 6.25% S; found: 49.30% C, 3.01% H, 2.69% N, 6.22% S; UV: 449

(3.32); $^1\text{H-NMR}$ (400 MHz): 8.52 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.29 (d, 1H, $J_{(5'',6'')} = 8.2$ Hz, H-6''), 8.33 (s, 1H, H-2''), 8.26 (d, 1H, $J_{(6,7)} = 8.2$ Hz, H-7), 8.17 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- β), 7.85 (t, 1H, $J_{(4,5)} = 8.0$ Hz, $J_{(5,6)} = 8.0$ Hz, H-6), 7.82 (t, 1H, $J_{(5,6)} = 8.0$ Hz, $J_{(6,7)} = 8.2$ Hz, H-5), 7.76 (d, 1H, $J_{(4'',5'')} = 7.9$ Hz, H-4''), 7.72 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- α) 7.69 (t, 1H, $J_{(4'',5'')} = 7.9$ Hz, $J_{(5'',6'')} = 8.2$ Hz, H-5''), 7.59 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.55 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 4.50 (s, 3H, $\text{N}^+\underline{\text{CH}_3}$); $^{13}\text{C-NMR}$ (100 MHz): 171.4 (C-2), 155.8 (C-5'), 151.0 (C-2'), 143.2 (C-3a), 134.1 (C- β), 130.9 (C-5''), 130.1 (CF_3), 129.5 (C-6), 128.9 (C-6''), 128.1 (C-5), 127.9 (C-7a), 126.1 (C-4''), 124.8 (C-7), 123.7 (C-3'), 122.2 (C-1''), 117.1 (C-4), 112.0 (C-4'), 110.5 (C- α), 37.1 ($\text{N}^+\underline{\text{CH}_3}$).

2-{(E)-2-[5-(2-Bromophenyl)furan-2-yl]vinyl}-3-methyl-1,3-benzothiazolium iodide (5g). Reaction time: 60 min; yield 60%; m.p. 215-219°C (ethanol); Calc. for $\text{C}_{20}\text{H}_{15}\text{BrINOS}$ (524.21): 45.82% C, 2.88% H, 15.24% Br, 2.67% N, 6.12% S; found: 45.96% C, 2.79% H, 15.31% Br, 2.59% N, 6.20% S; UV: 449 (3.44); $^1\text{H-NMR}$ (400 MHz): 8.62 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.44 (d, 1H, $J_{(6,7)} = 8.0$ Hz, H-7), 8.35 (d, 1H, $J_{(5'',6'')} = 7.8$ Hz, H-6''), 8.33 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- β), 8.06 (d, 1H, $J_{(4'',5'')} = 7.7$ Hz, H-5''), 7.97 (t, 1H, $J_{(3',4')} = 8.0$ Hz, $J_{(4'',5'')} = 7.7$ Hz, H-4''), 7.90 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- α), 7.77 (t, 1H, $J_{(4,5)} = 8.4$ Hz, $J_{(5,6)} = 7.7$ Hz, H-6), 7.72 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.69 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 7.59 (t, 1H, $J_{(5,6)} = 7.7$ Hz, $J_{(6,7)} = 8.0$ Hz, H-5) 4.53 (s, 3H, $\text{N}^+\underline{\text{CH}_3}$); $^{13}\text{C-NMR}$ (100 MHz): 171.2 (C-2), 155.6 (C-5'), 150.7 (C-2'), 142.4 (C-3a), 134.9 (C- β), 133.5 (C-5''), 131.3 (C-4''), 130.3 (C-6''), 129.8 (C-6), 129.4 (C-2''), 128.7 (C-3''), 128.6 (C-5), 128.1 (C-7a), 123.5 (C-7), 122.9 (C-3'), 120 (C-1''), 117.1 (C-4), 114.3 (C-4'), 111.2 (C- α), 36.7 ($\text{N}^+\underline{\text{CH}_3}$).

2-{(E)-2-[5-(2,4-Dichlorophenyl)furan-2-yl]vinyl}-3-methyl-1,3-benzothiazolium iodide (5h). Reaction time: 60 min; yield: 86 %; m.p. 248-253°C (ethanol); Calc. for $\text{C}_{20}\text{H}_{14}\text{Cl}_2\text{INOS}$ (514.21): 46.72% C, 2.74% H, 13.79% Cl, 24.68% I, 2.72% N, 3.11% O, 6.24% S; found: 46.72% C, 2.74% H, 13.79% Cl, 2.72% N, 6.24% S; UV: 449 (3.59); $^1\text{H-NMR}$ (400 MHz): 8.35 (d, 1H, $J_{(4,5)} = 8.4$ Hz, H-4), 8.30 (d, 1H, $J_{(5'',6'')} = 8.6$ Hz, H-6''), 8.25 (d, 1H, $J_{(6,7)} = 8.0$ Hz, H-7), 8.15 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- β), 7.88 (t, 1H, $J_{(4,5)} = 8.4$ Hz, $J_{(5,6)} = 7.6$ Hz, H-6), 7.84 (s, 1H, H-3''), 7.79 (t, 1H, $J_{(5,6)} = 7.6$ Hz, $J_{(6,7)} = 8.0$ Hz, H-5), 7.77 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- α), 7.64 (d, 1H, $J_{(5'',6'')} = 8.6$ Hz, H-5''), 7.53 (d, 1H, $J_{(3',4')} = 3.8$ Hz, H-3'), 7.50 (d, 1H, $J_{(3',4')} = 3.8$ Hz, H-4'), 4.36 (s, 3H, $\text{N}^+\underline{\text{CH}_3}$); $^{13}\text{C-NMR}$ (100 MHz): 170.7 (C-2), 152.6 (C-5'), 150.2 (C-2'), 141.9 (C-3a), 134.1 (C-4''), 132.8 (C- β), 130.7 (C-2''), 130.4 (C-5''), 130 (C-3''), 129.3 (C-6), 128.2 (C-6''), 127.8 (C-5), 127.7 (C-7a), 125.8 (C-1''), 124.1 (C-7), 122.5 (C-3'), 116.6 (C-4), 115.5 (C-4'), 111.1 (C- α), 36.2 ($\text{N}^+\underline{\text{CH}_3}$).

2-{(E)-2-[5-(4-Chlorophenyl)furan-2-yl]vinyl}-3-ethyl-1,3-benzothiazolium iodide (6a). Reaction time: 150 min; yield 62%; m.p. 245-246°C (methanol); Cal. for $\text{C}_{21}\text{H}_{17}\text{ClINOS}$ (493.79): 51.08% C, 3.47% H, 7.18% Cl, 2.84% N, 6.49% S; found: 51.74% C, 3.55% H, 7.21% Cl, 2.79% N, 6.53% S; UV 468 (3.61); $^1\text{H-NMR}$ (400 MHz): 8.43 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.28 (d, 1H, $J_{(6,7)} = 8.5$ Hz, H-7), 8.07 (d, 2H, $J_{(2'',3'')} = 8.6$ Hz, H-2'', H-6''), 8.14 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- β), 7.87 (t, 1H, $J_{(4,5)} = 8.0$ Hz, $J_{(5,6)} = 7.8$ Hz, H-6), 7.78 (t, 1H, $J_{(5,6)} = 7.8$ Hz, $J_{(6,7)} = 8.5$ Hz, H-5), 7.71 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- α),

7.61 (d, 2H, $J_{(2'',3'')} = 8.6$ Hz, H-3'', H-5''), 7.50 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.43 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 4.97 (q, 2H, N⁺CH₂), 1.49 (t, 3H, CH₃); ¹³C-NMR (100 MHz): 171.4 (C-2), 158.1 (C-5'), 151.4 (C-2'), 141.9 (C-3a), 135.1 (C-4''), 134.6 (C-β), 130.4 (C-6), 130.1 (C-2''), 129.2 (C-5), 129 (C-7a), 128.4 (C-1''), 127.7 (C-3''), 125.4 (C-7), 124.8 (C-3'), 117 (C-4), 112.1 (C-4'), 110.2 (C-α), 45.3 (N⁺CH₂), 15.1 (CH₃).

2-<{(E)-2-[5-(4-Bromophenyl)furan-2-yl]vinyl}-3-ethyl-1,3-benzothiazolium iodide (6b). Reaction time: 180 min; yield 45 %; m.p. 248-249°C (methanol); Calc. for C₂₁H₁₇BrINOS (538.24): 46.86% C, 3.18% H, 14.85% Br, 2.60% N, 5.96% S; found: 46.80% C, 3.08% H, 14.69% Br, 2.62% N, 5.82% S; UV 468 (3.61); ¹H-NMR (400 MHz): 8.43 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.27 (d, 1H, $J_{(6,7)} = 8.4$ Hz, H-7), 7.98 (d, 2H, $J_{(2'',3'')} = 8.6$ Hz, H-2'', H-6''), 8.11 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H-β), 7.85 (t, 1H, $J_{(5,6)} = 7.8$ Hz, $J_{(6,7)} = 8.4$ Hz, H-6), 7.76 (t, 1H, $J_{(4,5)} = 8.0$ Hz, $J_{(5,6)} = 7.8$ Hz, H-5), 7.71 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H-α), 7.70 (d, 2H, $J_{(2'',3'')} = 8.6$ Hz, H-3'', H-5''), 7.49 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.43 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 4.98 (q, 2H, N⁺CH₂), 1.47 (t, 3H, CH₃); ¹³C-NMR (100 MHz): 171.5 (C-2), 158.1 (C-5'), 151.5 (C-2'), 141.9 (C-3a), 134.6 (C-β), 133.0 (C-2''), 130.4 (C-6), 129.2 (C-5), 129 (C-7a), 128.7 (C-4''), 127.9 (C-3''), 125.4 (C-7), 124.8 (C-3'), 123.8 (C-1''), 117.4 (C-4), 112.1 (C-4'), 110.3 (C-α), 45.3 (N⁺CH₂), 15.1 (CH₃).

3-Ethyl-2-<{(E)-2-[5-(4-nitrophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium iodide (6c). Reaction time: 220 min; yield 79%; m.p. 265-268°C (ethanol); Calc. for C₂₁H₁₇IN₂O₃S (504.34): 50.01% C, 3.40% H, 5.55% N, 6.36% S; found: 50.08% C, 3.45% H, 5.61% N, 6.42% S; UV: 458 (3.57); ¹H-NMR (400 MHz): 8.46 (d, 1H, $J_{(4,5)} = 8.9$ Hz, H-4), 8.35 (d, 2H, $J_{(2'',3'')} = 8.9$ Hz, H-3'', H-5''), 8.29 (d, 2H, $J_{(2'',3'')} = 8.9$ Hz, H-2'', H-6''), 8.29 (d, 1H, $J_{(6,7)} = 8.9$ Hz, H-7), 8.17 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H-β), 7.88 (t, 1H, $J_{(4,5)} = 8.9$ Hz, $J_{(5,6)} = 7.6$ Hz, H-6), 7.80 (t, 1H, $J_{(5,6)} = 7.6$ Hz, $J_{(6,7)} = 8.9$ Hz, H-5), 7.71 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H-α), 7.66 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.55 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 5.01 (q, 2H, N⁺CH₂), 1.51 (t, 3H, CH₃); ¹³C-NMR (100 MHz): 170.3 (C-2), 155.3 (C-5'), 151.6 (C-2'), 146.9 (C-1''), 142.8 (C-3a), 134.1 (C-4''), 133.3 (C-β), 129.5 (C-6), 128.3 (C-5), 128.2 (C-7a), 125.6 (C-3''), 124.3 (C-2''), 124.3 (C-7), 123.1 (C-3'), 116.5 (C-4), 113.7 (C-4'), 110.7 (C-α), 44.4 (N⁺CH₂), 14.1 (CH₃).

3-Ethyl-2-<{(E)-2-[5-(3-nitrophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium iodide (6d). Reaction time: 90 min; yield 74%; m.p. 246-248°C (ethanol); Calc. for C₂₁H₁₇IN₂O₃S (504.34): 50.01% C, 3.40% H, 5.55% N, 6.36% S; found: 49.89% C, 3.35% H, 5.48% N, 6.41% S; UV: 449 (3.36); ¹H-NMR (400 MHz): 8.75 (s, 1H, H-2''), 8.49 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.43 (d, 1H, $J_{(5'',6'')} = 8.0$ Hz, H-6''), 8.31 (d, 1H, $J_{(4'',5'')} = 7.9$ Hz, H-4''), 8.27 (d, 1H, $J_{(6,7)} = 8.5$ Hz, H-7), 8.2 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H-β), 7.88 (t, 1H, $J_{(4,5)} = 8.0$ Hz, $J_{(5,6)} = 7.7$ Hz, H-6), 7.85 (t, 1H, $J_{(4'',5'')} = 7.9$ Hz, $J_{(5'',6'')} = 8.0$ Hz, H-5''), 7.80 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H-α), 7.80 (t, 1H, $J_{(5,6)} = 7.7$ Hz, $J_{(6,7)} = 8.5$ Hz, H-5), 7.66 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.55 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 4.99 (q, 2H, N⁺CH₂), 1.51 (t, 3H, CH₃); ¹³C-NMR (100 MHz): 170.5 (C-2), 155.4 (C-5'), 151.0 (C-2'), 148.5 (C-3''), 140.8 (C-3a), 133.5 (C-β), 130.7

(C-4''), 130.6 (C-1''), 129.4 (C-6), 128.2 (C-5), 128.1 (C-7a), 124.3 (C-5''), 123.5 (C-7), 123.0 (C-3''), 119.0 (C-2''), 116.5 (C-4), 112.4 (C-4'), 110.7 (C- α), 44.3 (N^+CH_2), 14.0 (CH_3).

3-Ethyl-2-{(E)-2-[5-(2-nitrophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium iodide (6e). Reaction time: 190 min; yield 46%; m.p. 227-231°C (ethanol); Calc. for $C_{21}H_{17}IN_2O_3S$ (504.34): 50.01% C, 3.40% H, 5.55% N, 6.36% S; found: 50.21% C, 3.42% H, 5.61% N, 6.41% S; UV: 439 (3.47); 1H -NMR (400 MHz): 8.45 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.32 (d, 1H, $J_{(6,7)} = 8.4$ Hz, H-7), 8.16 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- β), 8.1 (d, 1H, $J_{(5'',6'')} = 7.7$ Hz, H-6''), 8.01 (d, 1H, $J_{(3'',4'')} = 7.9$ Hz, H-3''), 7.88 (t, 1H, $J_{(4,5)} = 8.0$ Hz, $J_{(5,6)} = 7.7$ Hz, H-6), 7.84 (t, 1H, $J_{(4'',5'')} = 8.8$ Hz, $J_{(5'',6'')} = 7.7$ Hz, H-5''), 7.80 (t, 1H, $J_{(3'',4'')} = 7.8$ Hz, $J_{(4'',5'')} = 8.8$ Hz, H-4''), 7.72 (t, $J_{(5,6)} = 7.7$ Hz, $J_{(6,7)} = 8.4$ Hz, 1H, H-5), 7.50 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- α), 7.50 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.29 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 5.01 (q, 2H, N^+CH_2), 1.51 (t, 3H, CH_3); ^{13}C -NMR (100 MHz): 169.9 (C-2), 152.3 (C-5'), 151.2 (C-2'), 147.1 (C-2''), 140.8 (C-3a), 133.1 (C- β), 132.5 (C-5''), 130.7 (C-4''), 129.5 (C-6), 129.2 (C-6''), 128.3 (C-5), 128.2 (C-7a), 124.4 (C-3''), 124.0 (C-7), 122.0 (C-3'), 121.2 (C-1''), 116.4 (C-4), 113.6 (C-4'), 110.52 (C- α), 44.3 (N^+CH_2), 13.8 (CH_3).

3-Ethyl-2-{(E)-2-[5-[3-(trifluoromethyl)phenyl]furan-2-yl]vinyl}-1,3-benzothiazolium iodide (6f). Reaction time: 100 min; yield 58%; m.p. 170-174°C (methanol); Calc. for $C_{22}H_{17}F_3INOS$ (527.34): 50.11% C, 3.25% H, 2.66% N, 6.08% S; found: 50.22% C, 3.31% H, 2.72% N, 6.11% S; UV: 453 (3.24); 1H -NMR (400 MHz): 8.47 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.35 (d, 1H, $J_{(5'',6'')} = 7.6$ Hz, H-6''), 8.33 (s, 1H, H-2''), 8.31 (d, 1H, $J_{(6,7)} = 8.7$ Hz, H-7), 8.17 (d, 1H, $J_{(\alpha,\beta)} = 15.2$ Hz, H- β), 7.85 (t, 1H, $J_{(4,5)} = 8.0$ Hz, $J_{(5,6)} = 7.6$ Hz, H-6), 7.8 (t, 1H, $J_{(5,6)} = 7.6$ Hz, $J_{(6,7)} = 8.7$ Hz, H-5), 7.78 (d, 1H, $J_{(4'',5'')} = 7.7$ Hz, H-4''), 7.78 (d, 1H, $J_{(\alpha,\beta)} = 15.2$ Hz, H- α) 7.77 (t, 1H, $J_{(4',5')} = 7.7$ Hz, $J_{(5'',6'')} = 7.6$ Hz, H-5''), 7.61 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.55 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 4.99 (q, 2H, N^+CH_2), 1.50 (t, 3H, CH_3); ^{13}C -NMR (100 MHz): 170.6 (C-2), 156.3 (C-5'), 150.9 (C-2'), 141.0 (C-3a), 133.7 (C- β), 130.3 (C-5''), 129.9 (d, CF_3), 129.5 (C-6), 128.6 (C-6''), 128.3 (C-5), 128.2 (C-7a), 125.7 (C-4''), 124.4 (C-7), 123.4 (C-3'), 122.6 (C-1''), 116.5 (C-4), 112.0 (C-4'), 110.1 (C- α), 44.4 (N^+CH_2), 14.1 (CH_3).

2-{(E)-2-[5-(2-Bromophenyl)furan-2-yl]vinyl}-3-ethyl-1,3-benzothiazolium iodide (6g). Reaction time: 90 min; yield 47 %; m.p. 235-237°C (methanol); Calc. for $C_{21}H_{17}BrINOS$ (538.24): 46.86% C, 3.18% H, 15.24% Br, 2.67% N, 6.12% S; found: 46.95% C, 3.06% H, 15.32% Br, 2.74% N, 6.09% S; UV: 449 (3.65); 1H -NMR (400 MHz): 8.44 (d, 1H, $J_{(4,5)} = 7.9$ Hz, H-4), 8.30 (d, 1H, $J_{(6,7)} = 8.4$ Hz, H-7), 8.18 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- β), 8.1 (d, 1H, $J_{(5'',6'')} = 7.9$ Hz, H-6''), 7.84 (t, 1H, $J_{(4'',5'')} = 7.6$ Hz, $J_{(5'',6'')} = 7.9$ Hz, H-5''), 7.79 (t, 1H, $J_{(3',4')} = 8.0$ Hz, $J_{(4',5')} = 7.6$ Hz, H-4''), 7.70 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- α), 7.59 (t, 1H, $J_{(4,5)} = 7.9$ Hz, $J_{(5,6)} = 8.0$ Hz, H-6), 7.40 (t, 1H, $J_{(5,6)} = 8.0$ Hz, $J_{(6,7)} = 8.4$ Hz, H-5), 7.55 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.52 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 4.96 (q, 2H, N^+CH_2), 1.49 (t, 3H, CH_3); ^{13}C -NMR (100 MHz): 171.0 (C-2), 155.7 (C-5'), 150.7 (C-2'), 141.3 (C-3a), 133.8 (C- β), 132.5 (C-5''), 131.3 (C-4''), 130.7 (C-6''), 129.4 (C-6), 129.3 (C-2''), 128.7 (C-3''), 128.7 (C-7a),

128.6 (C-5), 124.8 (C-7), 122.9 (C-3'), 120.0 (C-1''), 116.9 (C-4), 115.1 (C-4'), 110.5 (C- α), 44.8 ($\text{N}^+\underline{\text{CH}_2}$), 14.5 ($\underline{\text{CH}_3}$).

2-{(E)-2-[5-(2,4-Dichlorophenyl)furan-2-yl]vinyl}-3-ethyl-1,3-benzothiazolium iodide (6h). Reaction time: 130 min; yield: 88 %, m.p. 260–263°C (methanol); Calc. for $\text{C}_{21}\text{H}_{16}\text{Cl}_2\text{INOS}$ (528.23): 47.75% C, 3.05% H, 13.42% Cl, 2.65% N, 6.07% S; found: 47.90% C, 3.15% H, 13.46% Cl, 2.71% N, 6.11% S; UV: 449 (3.59); $^1\text{H-NMR}$ (400 MHz): 8.44 (d, 1H, $J_{(4,5)} = 8.1$ Hz, H-4), 8.30 (d, 1H, $J_{(6,7)} = 8.5$ Hz, H-7), 8.27 (d, 1H, $J_{(5'',6'')} = 8.8$ Hz, H-6''), 8.17 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- β), 7.87 (t, 1H, $J_{(4,5)} = 8.1$ Hz, $J_{(5,6)} = 7.7$ Hz, H-6), 7.83 (s, 1H, H-3''), 7.80 (t, 1H, $J_{(5,6)} = 7.7$ Hz, $J_{(6,7)} = 8.5$ Hz, H-5), 7.75 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- α), 7.64 (d, 1H, $J_{(5'',6'')} = 8.8$ Hz, H-5''), 7.54 (br s, 2H, H-3', H-4'), 4.99 (q, 2H, $\text{N}^+\underline{\text{CH}_2}$), 1.48 (t, 3H, CH_3); $^{13}\text{C-NMR}$ (100 MHz): 170.4 (C-2), 152.9 (C-5'), 150.2 (C-2'), 140.8 (C-3a), 134.1 (C-4''), 133.4 (C- β), 130.8 (C-2''), 130.4 (C-5''), 130.1 (C-3''), 129.5 (C-6), 128.1 (C-6''), 128.1 (C-7a), 127.9 (C-5), 125.9 (C-1''), 124.3 (C-7), 122 (C-3'), 116.4 (C-4), 113.6 (C-4'), 110.5 (C- α), 44.3 ($\text{N}^+\underline{\text{CH}_2}$), 14 ($\underline{\text{CH}_3}$).

3-Benzyl-2-{(E)-2-[5-(4-chlorophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium bromide (7a). Reaction time: 70 min; yield: 91%; m.p. 232–235°C; Calc. for $\text{C}_{26}\text{H}_{19}\text{BrClNOS}$ (508.85): 61.37% C, 3.76% H, 15.70% Br, 6.97% Cl; found: 61.21% C, 3.63% H, 15.81% Br, 7.06% Cl; UV: 478 (3.96); $^1\text{H-NMR}$ (500 MHz): 8.55 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.24 (d, 1H, $J_{(6,7)} = 8.5$ Hz, H-7), 8.00 (d, 2H, $J_{(2'',3'')} = 8.6$ Hz, H-2'', H-6''), 8.26 (d, 1H, $J_{(\alpha,\beta)} = 15.3$ Hz, H- β), 7.82 (t, 1H, $J_{(4,5)} = 8.0$ Hz, $J_{(5,6)} = 7.8$ Hz, H-6), 7.84 (t, 1H, $J_{(5,6)} = 7.8$ Hz, $J_{(6,7)} = 8.5$ Hz, H-5), 7.93 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- α), 7.63 (d, 2H, $J_{(2'',3'')} = 8.6$ Hz, H-3'', H-5''), 7.58 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 7.47 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 6.34 (s, 2H, $\text{N}^+\underline{\text{CH}_2\text{Ph}}$), 7.40–7.44 (m, 4H, ArH); $^{13}\text{C-NMR}$ (125 MHz): 171.78 (C-2), 157.44 (C-5'), 150.59 (C-2'), 141.44 (C-3a), 134.29 (C-4''), 134.25 (C- β), 129.64 (C-6), 129.26 (C-3''), 128.13 (C-7a), 128.36 (C-5), 127.49 (C-1''), 126.81 (C-2''), 124.69 (C-4), 124.54 (C-4'), 116.89 (C-7), 111.38 (C-3'), 109.63 (C- α), 51.46 ($\text{N}^+\underline{\text{CH}_2\text{Ph}}$), 134.15, 129.26, 128.61, 127.11.

3-Benzyl-2-{(E)-2-[5-(4-bromophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium bromide (7b). Reaction time: 100 min; yield: 87%; m.p. 225–228°C; Calc. for $\text{C}_{26}\text{H}_{18}\text{Br}_2\text{NOS}$ (553.31): 56.44% C, 3.46% H, 28.88% Br, 2.53% N, 5.80% S; found: 56.62% C, 3.35% H, 29.21% Br, 2.74% N, 6.12% S; UV: 478 (3.67); $^1\text{H-NMR}$ (500 MHz): 8.55 (d, 1H, $J_{(4,5)} = 8.0$ Hz, H-4), 8.23 (d, 1H, $J_{(6,7)} = 8.4$ Hz, H-7), 8.32 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- β), 8.00 (d, 2H, $J_{(2'',3'')} = 8.6$ Hz, H-2'', H-6''), 7.86 (t, 1H, $J_{(4,5)} = 8.0$ Hz, $J_{(5,6)} = 7.8$ Hz, H-6), 7.82 (t, 1H, $J_{(5,6)} = 7.8$ Hz, $J_{(6,7)} = 8.4$ Hz, H-5), 7.79 (d, 2H, $J_{(2'',3'')} = 8.6$ Hz, H-3'', H-5''), 7.71 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- α), 7.56 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 7.48 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 6.34 (s, 2H, $\text{N}^+\underline{\text{CH}_2\text{Ph}}$); $^{13}\text{C-NMR}$ (125 MHz): 171.84 (C-2), 157.49 (C-5'), 150.59 (C-2'), 141.44 (C-3a), 134.26 (C- β), 132.27 (C-3''), 129.67 (C-6), 128.64 (C-5), 128.13 (C-7a), 127.84 (C-1''), 127.11 (C-2''), 124.65 (C-4), 124.50 (C-4'), 123.12 (C-4''), 116.89 (C-7), 114.43 (C-3'), 109.68 (C- α), 51.48 ($\text{N}^+\underline{\text{CH}_2\text{Ph}}$), 129.29, 128.72, 127.14.

3-Benzyl-2-{(E)-2-[5-(4-nitrophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium bromide (7c). Reaction time: 130 min; yield: 64%; m.p. 235–240°C; Calc. for C₂₆H₁₉BrN₂O₃S (519.41): 60.12% C, 3.69% H, 15.38% Br, 5.39% N, 6.17% S; found: 60.44% C, 3.45% H, 15.37% Br, 5.62% N, 6.22% S; UV: 463 (3.58); ¹H-NMR (500 MHz): 8.54 (d, 1H, J_(4,5) = 8.6 Hz, H-4), 8.40 (d, 2H, J_(2'',3'') = 8.9 Hz, H-3''), H-5''), 8.31 (d, 2H, J_(2'',3'') = 8.9 Hz, H-2''), H-6''), 8.27 (d, 1H, J_(6,7) = 8.6 Hz, H-7), 8.32 (d, 1H, J_(α,β) = 15.5 Hz, H-β), 7.88 (t, 1H, J_(4,5) = 8.9 Hz, J_(5,6) 7.6 Hz, H-5), 7.82 (t, 1H, J_(5,6) = 7.6 Hz, J_(6,7) = 8.6 Hz, H-6), 8.05 (d, 1H, J_(α,β) = 15.5 Hz, H-α), 7.71 (d, 1H, J_(3',4') = 3.7 Hz, H-4'), 7.60 (d, 1H, J_(3',4') = 3.7 Hz, H-3'), 6.38 (s, 2H, N⁺CH₂Ph); ¹³C-NMR (125 MHz): 171.85 (C-2), 155.78 (C-5'), 151.85 (C-2'), 147.27 (C-4''), 141.48 (C-3a), 134.35 (C-1''), 134.12 (C-β), 129.82 (C-6), 128.64 (C-5), 128.37 (C-7a), 125.84, (C-2''), 124.77 (C-4), 124.59 (C-3''), 117.09 (C-7), 123.85 (C-3'), 114.16 (C-4'), 111.2 (C-α), 51.66 (N⁺CH₂Ph), 129.34, 128.72, 127.18.

3-Benzyl-2-{(E)-2-[5-(3-nitrophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium bromide (7d). Reaction time: 100 min; yield: 86%; m.p. 245–247°C; Calc. for C₂₆H₁₉BrN₂O₃S (519.41): 60.12% C, 3.69% H, 15.38% Br, 5.39% N, 6.17% S; found: 60.13% C, 3.52% H, 15.43% Br, 5.56% N, 6.24% S; UV: 458 (3.73); ¹H-NMR (300 MHz): 8.74 (s, 1H, H-2''), 8.50 (d, 1H, J_(4,5) = 7.5 Hz, H-4), 8.44–8.48 (m, 3H, H_{arom}), 8.27 (d, 1H, J_(5'',6'') = 7.5 Hz, H-6''), 8.26 (d, 1H, J_(α,β) = 15.6 Hz, H-β), 7.94 (d, 1H, J_(α,β) = 15.6 Hz, H-α), 7.78–7.86 (m, 3H, H_{arom}), 7.66 (d, 1H, J_(3',4') = 3.6 Hz, H-3'), 7.42 (d, 1H, J_(3',4') = 3.6 Hz, H-4'), 7.37–7.39 (m, 4H, H_{arom}), 6.32 (s, 2H, CH₂Ph); ¹³C-NMR (75 MHz): 171.65 (C-2), 155.64 (C-5'), 151.0 (C-2'), 148.55 (C-3''), 141.37 (C-3a), 134.01 (C-β), 130.81 (C-6''), 130.08 (C-4''), 129.63 (C-1''), 128.15 (C-6), 127.00 (C-7a), 124.56 (C-7), 123.73 (C-5''), 123.65 (C-3'), 116.83 (C-4'), 110.56 (C-α), 51.4 (CH₂Ph), 128.55, 128.40, 119.06.

3-Benzyl-2-{(E)-2-[5-(2-nitrophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium bromide (7e). Reaction time: 75 min; yield: 68%; m.p. 194–198°C; Calc. for C₂₆H₁₉BrN₂O₃S (519.41): 60.12% C, 3.69% H, 15.38% Br, 5.39% N, 6.17% S; found: 60.32% C, 3.65% H, 15.47% Br, 5.61% N, 6.14% S; UV: 449 (3.62); ¹H-NMR (300 MHz): 8.49 (d, 1H, J_(4,5) = 8.5 Hz, H-4), 8.31 (d, 1H, J_(6,7) = 8.3 Hz, H-7), 8.21–8.29 (m, 5H, H_{arom}), 8.19 (d, 1H, J_(α,β) = 15.5 Hz, H-β), 7.98–8.06 (m, 6H, H_{arom}), 7.53 (d, 1H, J_(α,β) = 15.5 Hz, H-α), 7.39 (d, 1H, J_(3',4') = 3.7 Hz, H-3'), 7.24 (d, 1H, J_(3',4') = 3.7 Hz, H-4'), 6.20 (s, 2H, CH₂Ph); ¹³C-NMR (75 MHz): 171.23 (c-2), 154.98 (C-5'), 151.16 (C-2'), 149.11 (C-3''), 141.49 (C-3a), 134.11 (C-β), 130.91 (C-6''), 130.22 (C-4''), 129.68 (C-1''), 128.40 (C-6), 128.01 (C-7a), 124.70 (C-7), 123.33 (C-5''), 123.65 (C-3'), 116.78 (C-4'), 111.00 (C-α), 51.38 (CH₂Ph), 128.77, 128.70, 119.12.

3-Benzyl-2-{(E)-2-[5-[3-(trifluoromethyl)phenyl]furan-2-yl]vinyl}-1,3-benzothiazolium bromide (7f). Reaction time: 90 min; yield: 66%; m.p. 250–254°C; Calc. for C₂₇H₁₉BrF₃NOS (542.41): 59.79% C, 3.53% H, 14.73% Br, 2.58% N, 5.91% S; found: 60.10% C, 3.43% H, 14.86% Br, 2.64% N, 6.05% S; UV: 463 (3.58); ¹H-NMR (300 MHz): 8.49 (d, 1H, J_(4,5) = 8.1 Hz, H-4), 8.31 (d, 1H, J_(6,7) = 8.3 Hz, H-7), 8.21–8.29 (m, 5H, H_{arom}), 8.19 (d, 1H, J_(α,β) = 15.7 Hz, H-β), 7.98–8.06 (m, 6H, H_{arom}), 7.79 (d,

1H, $J_{(\alpha,\beta)} = 15.7$ Hz, H- α), 7.53 (d, 1H, $J_{(3',4')} = 3.6$ Hz, H-3'), 7.39 (d, 1H, $J_{(3',4')} = 3.6$ Hz, H-4'), 6.20 (s, 2H, $\underline{\text{CH}_2\text{Ph}}$); ^{13}C -NMR (75 MHz): 171.65 (C-2), 153.18 (C-5'), 150.29 (C-2'), 141.33 (C-3a), 133.91 (C- β), 130.83 (C-5'), 130.50 (C-4''), 130.03 (C-6''), 129.61 (C-6), 129.12 (C-2''), 128.49 (C-3''), 128.40 (C-7a), 128.16 (C-5), 128.81 (C-7), 124.58 (C-3'), 123.23 (C-1'), 116.83 (C-4), 115.36 (C-4'), 110.72 (C- α), 51.4 ($\underline{\text{CH}_2\text{Ph}}$), 134.28, 127.98, 126.92.

3-Benzyl-2-{(E)-2-[5-(2-bromophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium bromide (7g). Reaction time: 120 min; yield: 52%; m.p. 220-224°C; Calc. for $\text{C}_{26}\text{H}_{18}\text{Br}_2\text{NOS}$ (553.31): 56.44% C, 3.46% H, 28.88% Br, 2.53% N, 5.80% S; found: 56.65% C, 3.33% H, 29.07% Br, 2.84% N, 6.43% S; ^1H -NMR (300 MHz): 8.48 (d, 1H, $J_{(4,5)} = 8.2$ Hz, H-4), 8.28 (d, 1H, $J_{(6,7)} = 8.1$ Hz, H-7), 8.10-8.26 (m, 5H, H_{arom}), 8.08 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- β), 7.79 - 7.84 (m, 6H, H_{arom}), 7.62 (d, 1H, $J_{(\alpha,\beta)} = 15.5$ Hz, H- α), 7.59 (d, 1H, $J_{(3',4')} = 3.6$ Hz, H-3'), 7.53 (d, 1H, $J_{(3',4')} = 3.6$ Hz, H-4'), 6.27 (s, 2H, $\underline{\text{CH}_2\text{Ph}}$); ^{13}C -NMR (75 MHz): 171.68 (C-2), 155.12 (C-5'), 150.21 (C-2'), 141.36 (C-3a), 133.89 (C- β), 130.95 (C-5'), 129.82 (C-4''), 129.60 (C-6''), 129.01 (C-6), 128.79 (C-2''), 128.46 (C-3''), 128.38 (C-7a), 128.19 (C-5), 128.11 (C-7), 124.55 (C-3'), 123.11 (C-1'), 116.76 (C-4), 114.67 (C-4'), 110.24 (C- α), 51.33 ($\underline{\text{CH}_2\text{Ph}}$), 134.46, 134.23, 126.85.

3-Benzyl-2-{(E)-2-[5-(2,4-dichlorophenyl)furan-2-yl]vinyl}-1,3-benzothiazolium bromide (7h). Reaction time: 120 min; yield: 43%; m.p. 227-230°C; Calc. for $\text{C}_{26}\text{H}_{18}\text{BrCl}_2\text{NOS}$ (543.30): 57.48% C, 3.34% H, 13.05% Br, 14.71% Cl, 2.58% N, 5.90% S; found: 57.62% C, 3.23% H, 13.15% Br, 14.81% Cl, 2.64% N, 5.85% S; UV: 463 (3.59); ^1H -NMR (300 MHz): 8.48 (d, 1H, $J_{(4,5)} = 8.1$ Hz, H-4), 8.28 (d, 1H, $J_{(6,7)} = 8.1$ Hz, H-7), 8.10 - 8.26 (m, 5H, H_{arom}), 8.08 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- β), 7.78 - 8.07 (m, 6H, H_{arom}), 7.62 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- α), 7.59 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-3'), 7.52 (d, 1H, $J_{(3',4')} = 3.7$ Hz, H-4'), 6.27 (s, 2H, $\underline{\text{CH}_2\text{Ph}}$). δ_{C} (75 MHz): 171.65 (C-2), 153.18 (C-5'), 150.29 (C-2'), 141.33 (C-3a), 133.91 (C- β), 130.83 (C-5'), 130.03 (C-4''), 129.61 (C-6''), 129.12 (C-6), 128.49 (C-2''), 128.40 (C-3''), 128.16 (C-7a), 127.98 (C-5), 126.91 (C-7), 124.56 (C-3'), 123.23 (C-1'), 116.83 (C-4), 113.36 (C-4'), 110.71 (C- α), 51.40 ($\underline{\text{CH}_2\text{Ph}}$), 134.27, 130.49, 125.81.

2-{(E)-2-[5-(Methoxycarbonyl)-4H-furo[3,2-b]pyrrol-2-yl]vinyl}-3-methyl-1,3-benzothiazolium iodide (8a). Yield 61%; m.p. 288-292°C (methanol); Calc. for $\text{C}_{18}\text{H}_{15}\text{IN}_2\text{O}_3\text{S}$ (466.29): 46.36% C, 3.24% H, 6.01% N, 6.88% S; found: 46.51% C, 3.33% H, 6.03% N, 6.92% S. UV: 483 (3.66); ^1H -NMR (500 MHz): 12.24 (s, 1H, NH), 8.42 (d, 1H, H-7), 8.25 (d, 1H, H-4), 8.11 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- β), 7.88 (t, 1H, H-5), 7.79 (t, 1H, H-6), 7.63 (d, 1H, $J_{(\alpha,\beta)} = 15.4$ Hz, H- α), 7.48 (s, 1H, H-3'), 6.89 (s, 1H, H-6'), 4.32 (s, 3H, $\text{N}^+\underline{\text{CH}_3}$), 3.87 (s, 3H, CO_2CH_3); ^{13}C -NMR (125 MHz): 170.59 (C-2), 161.27 ($\underline{\text{CO}_2\text{CH}_3}$), 156.07 (C-2'), 151.54 (C-6'a), 142.26 (C-3a), 134.37 (C- β), 130.79 (C-3'a), 129.2 (C-5'), 128.49 (C-6), 127.82 (C-7a), 124.1 (C-7), 121.63 (C-5), 116.86 (C-4), 109.91 (C- α), 108.8 (C-3'), 95.65 (C-6'), 52.18 ($\text{CO}_2\underline{\text{CH}_3}$), 36.30 ($\text{N}^+\underline{\text{CH}_3}$).

*3-Benzyl-2-[(E)-2-[(5-(methoxycarbonyl)-4H-furo[3,2-*b*]pyrrol-2-yl)vinyl]-1,3-benzothiazolium bromide (8b).* Reaction time: 120 min; yield: 70%; m.p. 235–238°C; Calc. for C₂₄H₁₉BrN₂O₃S (495.39): 58.19% C, 3.87% H, 5.65% N, 6.47% S; found: 58.40% C, 3.72% H, 5.72% N, 6.54% S; UV: 497 (3.76); ¹H-NMR (500 MHz): 12.30 (s, 1H, NH), 8.51 (d, 1H, J_(4,5) = 8.2 Hz, H-4), 8.27 (d, 1H, J_(α,β) = 15.3 Hz, H-β), 8.20 (d, 1H, J_(6,7) = 8.1 Hz, H-4), 7.84 (t, 1H, H-6), 7.79 (t, 1H, H-5), 7.76 (d, 1H, J_(α,β) = 15.3 Hz, H-α), 7.60 (s, 1H, H-3'), 7.44–7.36 (m, 5H, H_{arom}), 6.86 (s, 1H, H-5'), 6.29 (s, 2H, N⁺CH₂), 3.87 (s, 3H, CO₂CH₃); ¹³C-NMR (125 MHz): 171.35 (C-2), 160.99 (C-4'), 155.92 (C-2'), 151.69 (C-6a'), 141.41 (C-3a), 135.43 (C-β), 134.52 (C-3a'), 129.63 (C-6), 129.54 (CO₂CH₃), 128.32 (C-5), 127.98 (C-7a), 124.67 (C-4), 116.75 (C-7), 109.38 (C-3'), 109.02 (C-α), 95.55 (C-5'), 51.98 (CO₂CH₃), 51.13 (N⁺CH₂), 133.98, 129.21, 128.47, 126.73.

*3-Benzyl-2-[(E)-2-[(5-(ethoxycarbonyl)-4-methylfuro[3,2-*b*]pyrrol-2-yl)vinyl]-1,3-benzothiazolium bromide (8c).* Reaction time: 110 min; yield: 48%; m.p. 223–226°C; Calc. for C₂₆H₂₃BrN₂O₃S (523.44): 59.66% C, 4.43% H, 5.35% N, 6.13% S; found: 59.84% C, 4.20% H, 5.27% N, 6.07% S; UV: 497 (3.83); ¹H-NMR (500 MHz): 8.51 (d, 1H, J_(4,5) = 8.3 Hz, H-4), 8.29 (d, 1H, J_(α,β) = 15.3 Hz, H-β), 8.21 (d, 1H, J_(6,7) = 8.2 Hz, H-7), 7.84 (t, 1H, H-6), 7.79 (t, 1H, H-5), 7.76 (d, 1H, J_(α,β) = 15.3 Hz, H-α), 7.69 (s, 1H, H-3'), 7.45–7.36 (m, 5H, H_{arom}), 6.83 (H-6'), 6.29 (s, 2H, N⁺CH₂), 4.31 (q, 2H, CO₂CH₂CH₃), 4.01 (NCH₃), 1.34 (t, 3H, CO₂CH₂CH₃); ¹³C-NMR (125 MHz): 171.36 (C-2), 160.56 (CO₂CH₂CH₃), 155.55 (C-2'), 149.13 (C-6a'), 141.38 (C-3a), 135.59 (C-β), 129.66 (C-6), 129.30 (C-4'), 133.94, 129.26, 128.50, 128.37 (C-5), 128.05 (C-7a), 124.60 (C-4), 116.72 (C-7), 109.36 (C-α), 108.86 (C-3'), 96.84 (C-5'), 60.59 (CO₂CH₂CH₃), 51.17 (N⁺CH₂), 35.08 (NCH₃), 14.30 (CO₂CH₂CH₃), 133.94, 129.26, 128.50, 126.77.

*3-Benzyl-2-[(E)-2-[(5-(ethoxycarbonyl)-4-benzylfuro[3,2-*b*]pyrrol-2-yl)vinyl]-1,3-benzothiazolium bromide (8d).* Reaction time: 120 min; yield: 67%; m.p. 242–245°C; Calc. for C₃₂H₂₇BrN₂O₃S (599.54): 64.11% C, 4.54% H, 4.67% N, 5.35% S; found: 64.28% C, 4.38% H, 4.72% N, 5.41% S; UV: 497 (3.98); ¹H-NMR (500 MHz): 8.49 (d, 1H, J_(4,5) = 8.1 Hz, H-4), 8.27 (d, 1H, J_(α,β) = 15.3 Hz, H-β), 8.21 (d, 1H, J_(6,7) = 8.1 Hz, H-7), 7.85 (t, 1H, H-6), 7.79 (t, 1H, H-5), 7.76 (d, 1H, J_(α,β) = 15.3 Hz, H-α), 7.52 (s, 1H, H-3'), 7.44–7.25 (m, 10H, H_{arom}), 6.99 (s, 1H, H-5'), 6.28 (N⁺CH₂), 5.78 (NCH₂), 4.31 (q, 2H, CO₂CH₂CH₃), 1.32 (t, 3H, CO₂CH₂CH₃); ¹³C-NMR (125 MHz): 171.51 (C-2), 160.59 (CO₂CH₂CH₃), 155.75 (C-2'), 149.46 (C-6a'), 141.44 (C-3a), 135.40 (C-β), 134.50 (C-3a'), 129.74 (C-6), 129.30 (C-4'), 128.42 (C-5), 116.81 (C-4), 109.82 (C-α), 108.98 (C-3'), 98.01(C-5'), 60.73 (CO₂CH₂CH₃), 51.26 (N⁺CH₂), 50.17 (NCH₂), 14.25 (CO₂CH₂CH₃), 137.68, 133.94, 129.25, 128.80, 128.54, 127.78, 127.10.

*3-Benzyl-2-[(E)-2-[(5-(methoxycarbonyl)-6-methoxymethylfuro[2,3-*b*]pyrrol-2-yl)vinyl]-1,3-benzothiazolium bromide (9).* Reaction time: 160 min; yield: 45%; m.p. 278–282°C; Calc. for C₂₆H₂₃BrN₂O₄S (539.44): 57.89% C, 4.30% H, 5.19% N, 5.94% S; found: 58.12% C, 4.18% H, 5.27% N, 5.95% S; ¹H-NMR (500 MHz): 8.48 (d, 1H, J_(4,5) = 8.3 Hz, H-4), 8.30 (d, 1H, J_(α,β) = 15.3 Hz, H-β),

8.22 (d, 1H, $J_{(6,7)} = 8.1$ Hz, H-7), 7.87 (t, 1H, H-6), 7.81 (t, 1H, H-6), 7.68 (d, 1H, $J_{(\alpha,\beta)} = 15.3$ Hz, H- α), 7.67 (s, 1H, H-4'), 7.42-7.37 (m, 5H, H_{arom}), 7.19 (s, 1H, H-3'), 6.26 (N^+CH_2), 5.83 (s, 2H, CH_2OCH_3), 3.87 (s, 3H, CO_2CH_3), 3.31 (s, 3H, CH_2OCH_3).

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