Determination of (2,3-epoxypropyl)trimethylammonium chloride, (3-chloro-2-hydroxypropyl)trimethylammonium chloride, and (2,3-dihydroxypropyl)trimethylammonium chloride by capillary electrophoresis

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Abstract

Capillary electrophoresis (CE) is a powerful technique to determine (2,3-epoxypropyl)trimethylammonium chloride, (3-chloro-2-hydroxy-propyl)trimethylammonium chloride and their hydrolysis product (2,3-dihydroxypropyl)trimethylammonium chloride in complex matrices. The separation was performed in a 180 mM copper(II) sulphate–4 mM formic acid buffer at pH 3. The UV inactive target compounds were detected by indirect UV detection at 215 nm with a reference wavelength of 300 nm—copper(II) being the chromophor. The results were compared to data acquired by ion-pair HPLC with perchlorate as ion pair former in a 1 M aqueous sodium perchlorate solution in 0.2 mM phosphoric acid (pH 3.4) on a RP 18 column and refractive index detection as reference method. Results of both methods are in good agreement.
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1. Introduction

(2,3-Epoxypropyl)trimethylammonium chloride (Quab 151, **1**) and (3-chloro-2-hydroxypropyl)trimethylammonium chloride (Quab 188, **2**) play an important role in the cationisation of starch produced as additive for the paper industry. The cationisation reaction can be performed in an alkaline slurry of the intact starch granules at moderate temperatures [1]. Under these conditions side reactions occur and (2,3-dihydroxypropyl)trimethylammonium chloride (Quab diol, **3**) is formed as the main by-product [2], which means a loss in reaction efficiency (cf. Fig. 1). The amount of linked cationic groups can be determined by elemental analysis of the starch derivatives, while the reagent and by-products are separated from the cationic starch by filtration. However, this purification is no longer possible when low-molecular-mass substances like alkylpolyglycosides (APG, **4**) with an average degree of polymerisation (DP) of 1.4 (DP 1–5) are

cationised. These substances are derived from starch and fatty alcohols as ecologically friendly detergents. Cation-isation establishes new properties and opens access to new applications for example for hair conditioners or textiles [3]. Therefore, it is very important to determine the residual amounts of Quab 188 (**2**) and the cancerogenic Quab 151 (**1**) [4] and the Quab diol (**3**), which is formed by hydrolysis of the reagents and is inefficient for the required cationisation of the APG (**4**). Determination of the active content of the reagent Quab 151 (**1**) is usually performed by titration of excessive HCl after conversion of the epoxide Quab 151 (**1**) to the chlorohydrin Quab 188 (**2**) [5]. The amount of Quab 188 (**2**) is obtained by the reversed reaction, the formation of the epoxide Quab 151 (**1**) with sodium hydroxide and back titration of excess of base [6]. Finally, the content of Quab diol (**3**) is determined iodometrically via the degree of oxidation [7]. However, these methods were not applicable to the APG–reagent mixture since the sugar also reacts. A number of publications report on the determination of the structurally related biologically active compound choline [8–13]. Most of these utilised ion chromatography—a technique suffering from the need of expensive ion exchange

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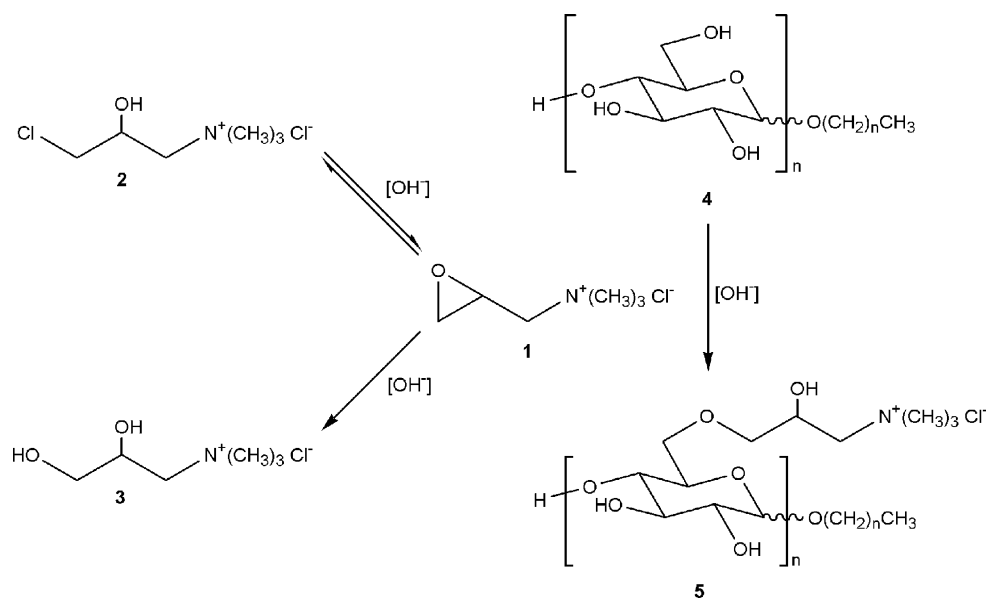*E-mail address:* p.mischnick@tu-bs.de (P. Mischnick).

Fig. 1. Reaction mechanism of the cationisation of alkylpolyglycosides (APG): Quab 151 (**1**), Quab 188 (**2**), Quab diol (**3**), alkylpolyglycoside (**4**), cationic alkylpolyglycoside (**5**).

columns, electrochemical detectors and repressor units for a sensitive detection of cations. The charged nature of the analytes offers a further way—capillary electrophoresis (CE). Literature about CE of cationic analytes preferably deals with the determination of inorganic cations, with cationic surfactants or pharmaceuticals. A method for the direct determination of the quaternary ammonium compounds **1**–**3** by CE with UV-Vis detection did not exist to the best of our knowledge.

Within our work on cationic APG (CA, **5**), a GC method for the determination of the degree of substitution (DS) and the substituent distribution in the glucosyl units of cationic APG (CA, **5**) was developed, which also allowed to calculate the reaction efficiency [14]. During our studies, Spruyt reported on the analysis of the hydrolysed cationic starches by ion-pair HPLC [15].

Our further goal was to find a method which allows the determination of remaining reagents and as far as possible cationic APG (CA, **5**) in one step. Since all compounds of interest are positively charged, and therefore not volatile as required for gas chromatography with its high separation efficiency, capillary zone electrophoresis (CZE) seemed to be a suitable method.

2. Experimental

2.1. General

The cationic APG (CA 1–9, **5**), Quab 151 (**1**) and Quab 188 (**2**) were obtained from Cognis Deutschland, Düsseldorf, Germany. The ratio of anhydro glucosyl units (AGU) to NaOH and Quab 151 (**1**) or the corresponding Quab 188

(**2**) were varied. Samples and data are listed in Table 1. Both standards, **1** and **2**, contained the hydrolysis product Quab diol (**3**). Copper(II) sulphate, 18-crown-6 (1,4,7,10,13,16-hexaoxadecane), ethyl alcohol, formic acid, hydrochloric acid, magnesium chloride, methanol, phenolphthalein, phosphoric acid (pK_a = 2.5), potato starch, sodium arsenite, sodium hydroxide and sodium perchlorate were of highest purity available and purchased from Fluka, Aldrich or Merck. For CE and HPLC water of NANOpure quality (pH 5.5) was used. The pH was adjusted with a pH meter 691 (Metrohm, Herisau, Switzerland) which was calibrated with buffers at pH 4 and 7.

Table 1
Structural features and reaction conditions for the synthesis of cationic APG (**5**)

Cationic APG	DP[a]	Alkyl chain	Molar ratio AGU[b]:Quab[c]:NaOH	DS[d]
CA 1	1	$C_{12/14}$	1:1.44:i.n.a.[e]	0.10
CA 2	1	C_{12}	1:1:0.05	0.17
CA 3	1	C_{12}	1:2.5:0.05	0.11
CA 4	1	C_{12}	1:5:0.05	0.17
CA 5	1	C_{12}	1:1:1.1	0.30
CA 6	1–5	C_{12}	1:1:0.28	0.12
CA 7	1–3	2-Ethylhexyl	1:2.9:0.05	0.53
CA 8[f]	1–3	2-Ethylhexyl	1:2.9:0.05	0.67
CA 9	1–3	2-Ethylhexyl	1:5:i.n.a.	0.52

[a] Degree of polymerisation.
[b] AGU: anhydro glucose unit.
[c] Quab: Quab 151 (CA 1–4, 7–9) respectively. Quab 188 (CA 5–6).
[d] Degree of substitution determined by GLC after dealkylation with morpholine [14].
[e] Information not available.
[f] Purified by membrane filtration.

2.2. Capillary electrophoresis

The electropherograms were generated using a P/ACE MDQ system (Beckman-Coulter, Munich, Germany). Fused-silica capillaries (Beckman-Coulter, München, Germany) of 50 μm inner diameter and 57 cm total length were used. The detection window was located 10 cm from the end of the capillary. Injections were performed hydrodynamically for 5 s with 40 mbar. To obtain positive peaks when using indirect UV detection, the polarity of the photodiode-array detection (DAD) system was reversed. Wavelength setting was 300 nm. The reference wavelength was set to 215 nm. Voltage: 20 kV, separation temperature: 25 °C, used computer program (Beckman-Coulter): P/ACE MDQ "Software Version 1.5".

2.2.1. Buffer solutions for CE

Copper(II) sulphate (4–180 mM), formic acid (4–80 mM), methanol (5–30%, v/v), 18-crown-6 (4 mM); optimised conditions: 180 mM copper(II) sulphate, 4 mM formic acid, pH 3 (adjusted with NaOH).

2.2.2. Rinsing procedures

New capillaries were rinsed as follows: water (10 min), 1 M HCl (10 min), water (2 min), 1 M NaOH (20 min), 0.1 M NaOH (20 min), water (30 min), each procedure with 2000 mbar; 45 min 30 kV. The procedure before each measurement was: water (5 min), buffer (15 min), each procedure with 2000 mbar; rinsing procedure at the end of the day: water (20 min), MeOH (10 min), 1 M HCl (10 min), water (5 min), 1 M NaOH (20 min), water (1 min), 0.1 M NaOH (10 min), water (10 min), each procedure with 2000 mbar.

2.2.3. Calibration

The CE system was calibrated with mixed standard solutions of Quab 151 (**1**) and Quab 188 (**2**). The content of **1** and **2** in the stock solution was determined by acidimetric titration and the content of **3** iodometrically via the degree of oxidation.

2.2.4. Sample preparation

The samples were dissolved in NANOpure water and ultrafiltrated.

2.3. Titrimetric determinations

The content of epoxide Quab 151 (**1**) was determined by titration of excessive HCl with 0.25 M NaOH after conversion with concentrated HCl and magnesium chloride to the chlorohydrin Quab 188 (**2**) [5]. The content of chlorohydrin Quab 188 (**2**) was determined by the reversed reaction with sodium hydroxide and back titration of excess of base with 1 M HCl. Phenolphthalein (0.1%, w/w, in ethanol) was used as the indicator [6].

Quab diol (**3**) was oxidised with 0.005 M sodium periodate. An excess of 0.005 M sodium arsenite was added to reduce remaining periodate whereas arsenite is oxidised to arsenate. The residual amount of arsenite was determined by iodometric titration. Potato starch solution (1%, w/w, in water) was added for endpoint determination [7].

2.4. Ion-pair HPLC

Ion-pair HPLC was performed according to the method of Spruyt [15] at room temperature using a Merck–Hitachi system consisting of a Merck–Hitachi L-6000 Pump, a Merck differential refractometer RI-71 and a Merck–Hitachi D-2500 Chromato-Integrator (Merck, Darmstadt, Germany). An Alltima C_{18} 5 U column (250 mm × 4.6 mm, 5 mm) was used. The injection volume was 20 μl. Isocratic eluent: 1 M sodium perchlorate in 0.2 mM phosphoric acid (pH 3.4) at a flow rate of 1 ml/min.

2.4.1. Calibration

The HPLC system was calibrated with mixed standard solutions of Quab 151 (**1**) and Quab 188 (**2**). The content of technical **1** and **2** was determined by acidimetric titration and the content of **3** iodometrically via the degree of oxidation.

2.4.2. Sample preparation

The samples were dissolved in NANOpure and water ultrafiltrated.

2.5. CE–electrospray ionisation (ESI)-MS

CE–ESI-MS was performed with a P/ACE 5500 system (Beckman-Coulter). A fused-silica capillary (Beckman-Coulter) of 50 μm inner diameter and 80 cm total length was used. Injections were performed hydrodynamically for 5 s with 40 mbar without any back pressure. The CE was coupled to the ESI mass spectrometer Esquire (Bruker Daltonik, Bremen, Germany), equipped with an ion trap. Mode: ESI—positive, drying gas: helium 4 l/min, temperature: 325 °C, capillary: −3500 V, end plate offset: −500 V, nebulizer: 10 p.s.i. (1 p.s.i. = 6894.76 Pa), cap exit: 120 V, cap exit offset: 90 V, skim I: 30 V, skim II: 10 V, trap drive: 50.4, cut-off: 55 *m/z*, scan range: 100–1000 *m/z*.

2.5.1. Buffer solution for CE-ESI/MS

4 mM Copper(II) sulphate, 4 mM formic acid, 30% methanol, pH 3 (adjusted with NaOH).

3. Results and discussion

The aim of our work was to develop an appropriate method for the simultaneous determination of cationic APG (CA, **5**, cf. Section 2 and Table 1) and remaining reagents from a raw mixture of cationised alkylpolyglucosides (cf. Fig. 1). Under alkaline conditions Quab 151 (**1**) and Quab 188 (**2**) were expected to be completely hydrolysed to the Quab diol (**3**) [3]. Therefore, the determination of Quab

diol (**3**) seemed to be suitable in order to control reaction efficiency. A very common method is the periodate oxidation where formaldehyde is released [16,17]. Cationic APG (**5**) should not disturb this approach since depending on the positions of the glucosidic linkages they should give either dialdehyde structures (1,4-linked or 1,2-linked), or formic acid (1,6-linked), or even no cleavage (1,3-linked). Formaldehyde was determined photometrically after reaction with 2,4-pentadione and ammonia as lutidine [18] and as 2,4-dinitro-phenylhydrazone by HPLC–UV [19]. While calibration of both methods using ethylene glycol under the same conditions showed a good reliability, the results obtained from the raw cationic APG samples were not plausible, but too high, especially from the lutidine method. Side products were also observed by HPLC–ESI-MS of the 2,4-dinitrophenylhydrazones, but the reason of interference could not be clarified. From these results it was obvious that a direct approach instead of an indirect one was required for these complex sample mixtures. Moreover, ^{1}H NMR and ESI-MS proved the existence of residual Quab 151 (**1**) and Quab 188 (**2**) [14].

3.1. Capillary electrophoresis

Riviello and Harrold [20] described the indirect detection of alkali cations in a buffer system consisting of 4 mM copper sulphate, 4 mM formic acid and 4 mM 18-crown-6. Copper(II) ions form solely $[Cu(H_2O)_4]^{2+} \cdot 2H_2O$ complexes with an absorption maximum at 215 nm in acidic aqueous solutions. Therefore, it can be used as background chromophor. The acidic medium (pH 3) is important to prevent dropping out of copper hydroxide which would clog the capillary. The crown ether which was used by the authors to separate potassium and ammonia could be omitted for our purpose. Its inner core has a size of 1.3–1.6 Å, and therefore it only complexes with cations, which are of appropriate size, e.g. potassium (1.52 Å), will be formed. Copper(II) is too small (0.87 Å) for any interactions [21]. An electropherogram received under the conditions of Riviello and Harrold is shown in Fig. 2. For peak assignment, standards of **1** and **2**, containing the hydrolysis product **3**, were used. As can be seen in Fig. 2, sodium migrates the fastest, followed by Quab 151 (**1**), some unidentified peaks, possibly addition products of **1**, and only one signal for Quab 188 (**2**) and Quab diol (**3**). The peaks are strongly distorted which is typical for CE separations with a wide range of electrophoretic mobilities of the analytes and indirect detection, where the peak form depends on a good matching between the mobility of the analyte and that of the background electrolyte. Analytes with higher mobility appear as peaks with a fronting, while analytes with lower mobility show a tailing. To achieve a separation of the Quab 188 (**2**) and the Quab diol (**3**), methanol was added to the buffer solution due to the very good solubility of the samples in this solvent. Moreover, it is used to increase the solvent strength, and minimises the hydrophobic interactions between the analytes and the capillary wall,
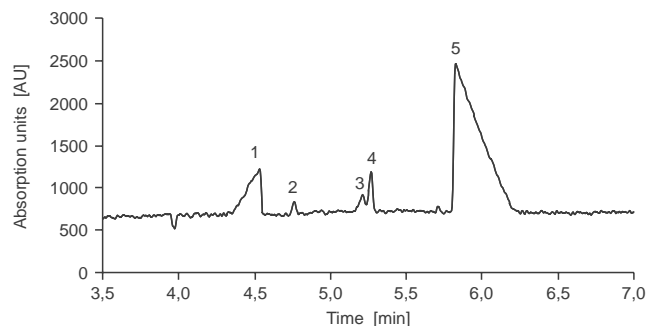

Fig. 2. Electropherogram of CA 1. Buffer [20]: [4 mM copper(II) sulphate, 4 mM formic acid, 4 mM 18-crown-6, pH 3]; 1: Na$^+$, 2: Quab 151 (**1**), 3 and 4: unknown, 5: Quab 188 (**2**) + Quab diol (**3**).

which occur under acidic conditions [22,23]. A stepwise increase of the methanol content up to 30% leads to baseline separation of the signal. But the high volatility of methanol caused imprecise quantitative results in the CE system with open sample and buffer vials. Another disadvantage is the deterioration of the baseline with increasing methanol content (cf. Fig. 3). Lowering the pH from 3.0 to 2.4 by increasing the formic acid concentration from 4 to 80 mM effected no improvement of separation, but only baseline drift and reduced response (not shown). A better way to optimise the separation was found by enhancing the copper sulphate concentration. This increase of ionic strength causes a decrease of the electroosmotic flow (EOF) and as a result a deceleration of the analytes migrating to the cathode. Fig. 4 illustrates the changes while varying the concentration from
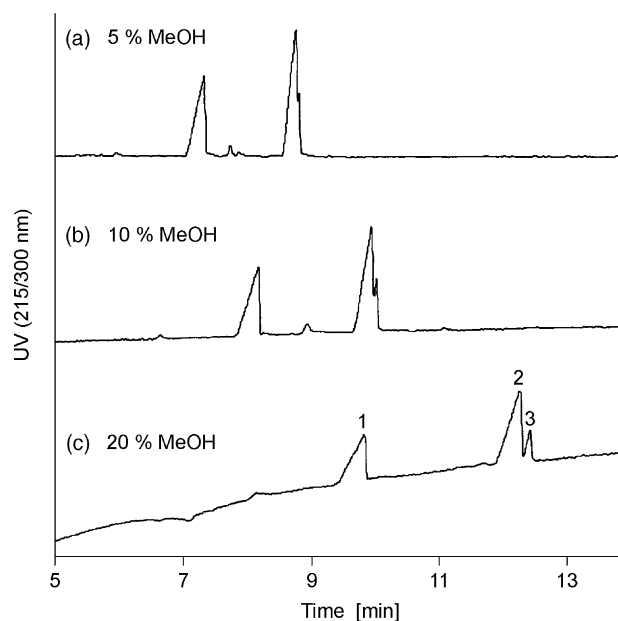

Fig. 3. Influence of methanol as modifier on the CE separation of a mixed standard of Quab compounds **1**–**3**. Buffer: 4 mM copper(II) sulphate, 4 mM formic acid, pH 3, methanol: (a) 5%, (b) 10% and (c) 20%; 1: Quab 151 (**1**), 2: Quab 188 (**2**), 3: Quab diol (**3**).
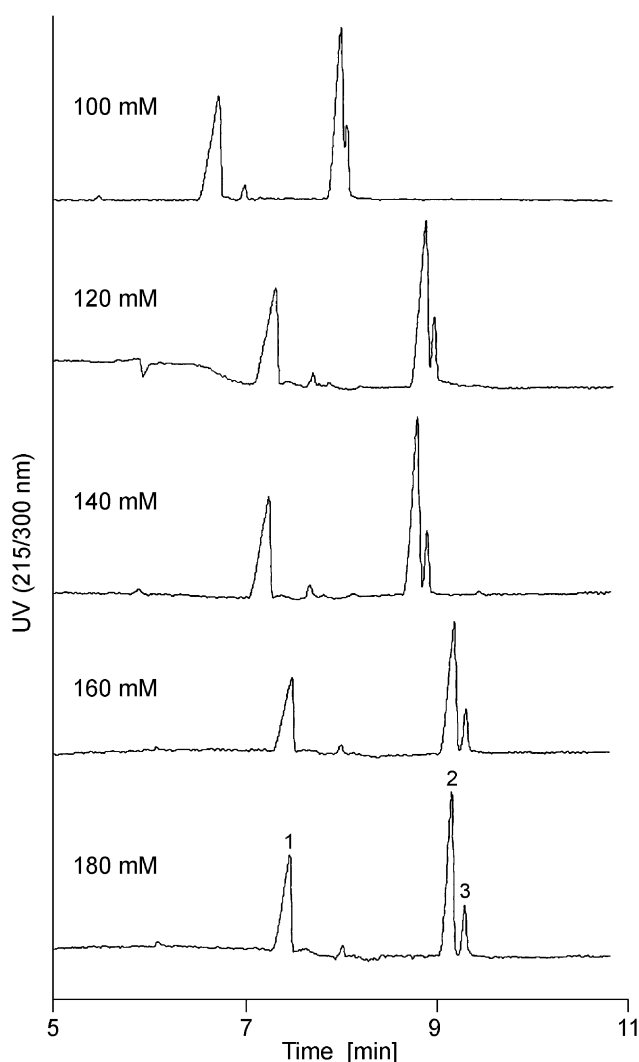
Fig. 4. Influence of the copper(II) sulphate concentration on the CE separation of a mixed standard of Quab compounds **1–3**. Buffer: 100–180 mM copper(II) sulphate, 4 mM formic acid, pH 3; 1: Quab 151 (**1**), 2: Quab 188 (**2**), 3: Quab diol (**3**).
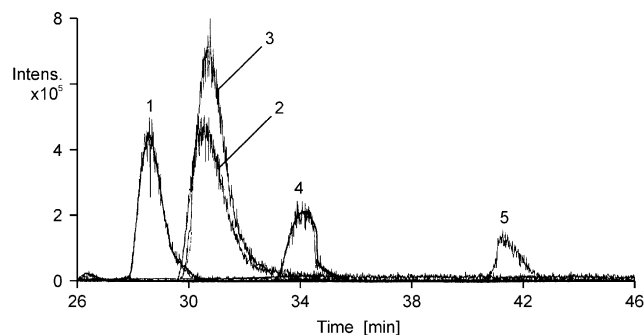

Fig. 5. Ion current chromatogram of CA 3 recorded by CE–ESI-MS. Buffer: 4 mM copper(II) sulphate, 4 mM formic acid, pH 3, 30% methanol; peaks were detected by m/z traces: 1: m/z 116 = Quab 151 m/z (**1**), 2: m/z 152 = Quab 188 (**2**), 3: m/z 134 = Quab diol (**3**), 4: m/z 290 = APG-C$_{12}$ disubstituted.; 5: m/z 464 = APG-C$_{12}$, monosubstituted.

4 to 180 mM copper(II) sulphate. At 180 mM a baseline separation was achieved. Higher concentrations were not considered to prevent problems with precipitation inside the capillary. Surprisingly, the cationic APG (**5**) could not be detected in the electropherogram. Bazzanella and Bächmann [24] found the same absorption of $[Cu(H_2O)_4]^{2+} \cdot 2H_2O$ before and after addition of various sugar compounds at pH 5, indicating that there is no complex formation under acidic conditions. This is a good prerequisite for indirect detection, since displacement of copper ions by the analyte causes a decrease of the background absorption. Until now it could not be clarified why the cationic APG (**5**) are not detected in our system. In contrast, by CE–ESI-MS measurements of CA 3 (APG-C$_{12}$, DP1, DS ca. 0.1) it could be shown that the cationic APG (**5**) are eluted shortly after the reagents. The CE conditions applied were not optimal, but only 4 mM copper(II) sulphate and 30% (v/v) methanol

were used since higher electrolyte concentrations are not compatible with electrospray (cf. Fig. 5). In addition to this sub-optimal buffer composition coupling was only possible with a transfer capillary of 80 cm length which could not be cooled. Under these conditions migration times strongly increased and peaks were broadened compared to the CE measurements and not reproducible. Cationic APG (**5**) could now be assigned by their m/z traces. At the acidic pH the EOF can be neglected and the analytes are eluted in the order of decreasing charge/mass ratio: Quab 151 (**1**) first, followed by co-eluting Quab 188 (**2**) and Quab diol (**3**), then the double charged disubstituted dodecylglucoside and finally the monosubstituted glucoside, as expected.

3.2. Quantitative analysis

The CE–UV method was applied to the quantitative analysis of the cationic reagents **1**, **2** and **3** in the cationic APG samples (CA 1–9). Peak areas were corrected for their various migration times and related to the concentration. Equations: (**1**) $y = 603.82x - 7.0149$, $R^2 = 0.996$; (**2**) $y = 572.63x + 63.624$, $R^2 = 0.997$; (**3**) $y = 917.93x - 8.463$, $R^2 = 0.993$. Calibration with standard solutions showed linearity in the concentration range tested: 70–13,000 μg/ml for Quab 151 (**1**), 70–10,000 μg/ml for Quab 188 (**2**) and 6–1500 μg/ml for Quab diol (**3**). Limit of detection was (**1**) 35 μg/ml, (**2**) 20 μg/ml and (**3**) 40 μg/ml. The limits of determination were (**1**) 50 μg/ml, (**2**) 35 μg/ml, and (**3**) 50 μg/ml. Results are shown in Table 2.

3.3. Ion-pair HPLC

As a reference method ion-pair HPLC was applied. The method uses a RP18 column and a sodium perchlorate solution adjusted to pH 3.4 with phosphoric acid as eluent. Perchlorate forms ion pairs with the cationic analytes. At the acidic pH of the eluent the very strong perchloric acid still is completely dissociated ($pK_a = -10$ [21]).

Table 2
Determination of Quab 151 (**1**), Quab 188 (**2**) and Quabdiol (**3**) by capillary electrophoresis [180 mM copper(II) sulphate, 4 mM formic acid, pH 3] and ion-pair chromatography (1 M sodium perchlorate + 0.2 mM μl/l phosphoric acid, pH 3.4)

Cationic APG	Quab 151 (g/100 g)		Quab 188 (g/100 g)		Quab diol (g/100 g)	
	CE	IPC	CE	IPC	CE	IPC
CA 1	n. d.[a]	n. d.	n. d.	n. d.	13.2	17.8
CA 2	2.1	2.4	1.7	2.5	12.3	13.1
CA 3	16.4	20.5	2.2	4.5	11.0	12.6
CA 4	14.7	21.5	11.9	24.8	19.2	23.0
CA 5	0.8	2.0	n. d.	1.6	6.5	7.1
CA 6	n. d.	1.1	n. d.	n. d.	2.3	2.6
CA 7	10.9	12.4	8.7	9.2	9.2	10.8
CA 8[b]	n. d.	n. d.	n. d.	n. d.	n. d.	n. d.
CA 9	12.6	13.2	40.5	37.4	10.5	9.7

For details see under Section 2. Values describe the mean of 2–5 measurements.

[a] n.d.: not detected.

[b] CA 8 was purified by ultrafiltration.

The ion pairs show the retention behaviour of uncharged species and elute in the order of decreasing polarity, i.e. Quab diol (**3**) before Quab 151 (**1**) and Quab 188 (**2**) (cf. Fig. 6). A second peak close behind Quab 151 (**1**) is probably caused by already mentioned oligomers of that compound.

Peak areas were related to the concentration. Equations: (**1**) $y = 312743x - 79643$, $R^2 = 0.9998$; (**2**) $y = 367917x - 61097$, $R^2 = 0.9994$; (**3**) $y = 446959x - 16351$, $R^2 = 0.9947$. Calibrations were performed in the concentration range 700–7000 μg/ml for Quab 151 (**1**), 70–7000 μg/ml for Quab 188 (**2**) and 6–600 μg/ml for Quab diol (**3**). Limit of detection was (**1**) 35 μg/ml, (**2**) 20 μg/ml and (**3**) 40 μg/ml. The limits of determination were (**1**) 50 μg/ml, (**2**) 35 μg/ml and (**3**) 50 μg/ml. The results are presented in Table 2.
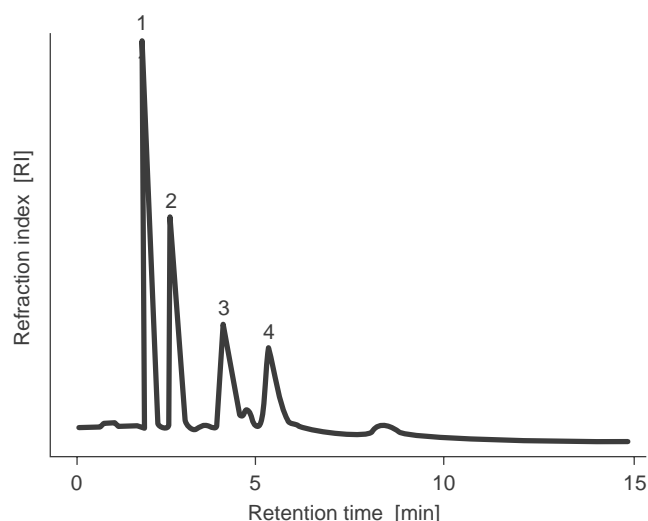

Fig. 6. Ion-pair chromatogram of a mixed standard of Quab compounds on a RP 18 column; eluent: 1 M sodium perchlorate + 0.2 mM phosphoric acid (pH 3.4); 1: injection peak, 2: Quab diol (**3**), 3: Quab 151 (**1**), 4: Quab 188 (**2**).

4. Discussion

The results obtained from capillary electrophoresis are in good correspondence with the results obtained from ion-pair chromatography. The estimated limits of detection and determination are comparable (cf. Section 2). Some of the samples (CA 3, 4, 7 and 9) show high amounts of cationic reagents. This is not very amazing because we performed the measurements with technical samples which were not purified, but only neutralised after the reaction time with the exception of CA 8 which was purified by ultrafiltration. Where a high excess of Quab was used as listed in Table 1, reaction efficiency was very poor [14], and residual amounts of **1**, **2** and **3** were high (CA 3, 4, 7, and 9). CA 1 indicates that all active Quab 151 (**1**) and Quab 188 (**2**) can be hydrolysed under appropriate reaction conditions. It is evident that the results for CA 4 vary a lot. The reason for this deviation is the character of the sample which was very difficult to handle due to its brittle and heterogeneous consistency. This applies also to CA 3.

5. Conclusion

Capillary electrophoresis with indirect UV detection using a copper sulphate/formic acid buffer is a suitable method for the determination of (2,3-epoxypropyl)trimethylammonium (Quab 151, **1**), (3-chloro-2-hydroxypropyl)trimethylammonium (Quab 188, **2**) and (2,3-dihydroxypropyl)trimethylammonium (Quab diol, **3**) compounds in complex matrices. The results were in good agreement with those obtained by ion-pair HPLC. Although CE requires buffer solutions, electrolyte concentration can be chosen much lower than in ion-pair HPLC and therefore enables coupling with electrospray ionisation mass spectrometry. While the cationic APG (**5**) were not eluted from the RP phase, they could be detected by CE–ESI-MS.

Acknowledgements

References

[1] O.B. Wurzburg (Ed.), Modified Starches: Properties and Uses, CRC Press, Boca Raton, FL, 1987.

[2] G. Hellwig, D. Bischoff, A. Rubo, Starch/Stärke 44 (1992) 69.

[3] K. Hill, W.V. Rybinski, G. Stoll, R. Tunn, Alkyl Polyglycosides, VCH, Weinheim, 1997.

[4] Technische Regeln für Gefahrstoffe: TRGS 905—Verzeichnis krebserzeugender, erbgutverändernder oder fortpflanzungsgefährdender Stoffe, Ausgabe März, 2001.

[5] Degussa AG: Analysenvorschrift zur Bestimmung des Epoxidgehaltes in Quab 151, Frankfurt, 1994.

[6] Analysenvorschrift zur Bestimmung des Chlorhydringehaltes in Quab 151, Degussa, Frankfurt, 1994.

[7] Analysenvorschrift zur Bestimmung des Glykolgehaltes in Quab 151, Degussa, Frankfurt, 1995.

[8] E. Haen, H. Hagenmaier, J. Remien, J. Chromatogr. 537 (1991) 514.

[9] D.N. Buchanan, F.R. Fucek, E.F. Domino, J. Chromatogr. 181 (1980) 329.

[10] J. Gorham, J. Chromatogr. 362 (1986) 243.

[11] J. Gorham, E. McDonnell, J. Chromatogr. 350 (1985) 245.

[12] L.D. Acevedo, Y. Xu, X. Zhang, R.J. Pearce, A. Yergey, J. Mass Spectrom. 31 (1996) 1399.

[13] K.-G. Wahlund, A. Sokolowski, J. Chromatogr. 151 (1978) 299.

[14] V. Goclik, P. Mischnick, Carbohydr. Res. 338 (2003) 733.

[15] D. Spruyt, in: Presented at the 54th Starch Congress, Detmold, 2003.

[16] Deutsche Gesellschaft für Fettwissenschaft, DGF (Ed.), Deutsche Einheitsmethoden zur Untersuchung von Fetten, Fettprodukten, Tensiden und verwandten Stoffen: E-III 3b (79), Wiss.-Verl.-Ges., Stuttgart, 2. Aufl., Loseblatt-Ausgabe, 1992.

[17] Bundesgesundheitsamt (Ed.), Amtliche Sammlung von Untersuchungsverfahren nach § 35 LMBG: K 84.00 7 (EG), Beuth, Berlin, Loseblatt-Ausgabe, 1982.

[18] R.L. Whistler, M.L. Wolfrom, J.N. BeMiller (Eds.), Methods in Carbohydrate Chemistry, Academic Press, New York, 1962, p. 441.

[19] J. Houben, T. Weyl (Eds.), Methoden der organischen Chemie, IV/1a, fourth ed., Thieme, Stuttgart, 1981, Chapter A 1, p. 450.

[20] J.M. Riviello, M.P. Harrold, J. Chromatogr. A 652 (1993) 385.

[21] A.F. Holleman, E. Wiberg (Eds.), Lehrbuch der Anorganischen Chemie, 101st ed., Walter de Gruyter, Berlin/New York, 1995, p. 238.

[22] H. Engelhardt, W. Beck, T. Schmitt (Eds.), Kapillarelektrophorese, Vieweg, Braunschweig, Wiesbaden, 1994.

[23] P. Britz-Mckibbin, D.D.Y. Chen, Anal. Chem. 70 (1998) 907.

[24] A. Bazzanella, K. Bächmann, J. Chromatogr. A 799 (1998) 283.