Journal of Dental Sciences (2019) 14, 346–351

Available online at www.sciencedirect.com

ScienceDirect

journal homepage: www.e-jds.com

Original Article

Solubility of dental core build-up materials in electric fields

Venus Hassani Katehsari*, Wilhelm Niedermeier **

Department of Prosthetic Dentistry, Center and School of Oral Medicine, University of Cologne, Cologne, Germany

Received 19 December 2018; Final revision received 29 March 2019
Available online 30 May 2019

KEYWORDS
Cement;
Composite;
Dental materials;
Oral galvanism;
Solubility

Abstract *Background/purpose:* Over 250 years has been researched over the consequences of oral galvanism. Previous studies have already examined the influence on dental fixation materials. The aim of this study was to investigate the electro-chemical solubility behavior of different dental core build-up materials in a pseudo-realistic galvanic experimental setup.
Materials and methods: The composite Admira (Voco, Cuxhaven, Germany) and two glass ionomer cements, Ketac Molar and Photac (3M-Espe, Seefeld, Germany), were examined. Test specimens were exposed to electric field strengths of 10–27 V/m in 0.9% saline solution. After 1, 2, 3, and 24 h, 2 ml of the electrolyte was removed for analysis. Aluminum and calcium were selected as parameters to measure the solubility of the products. Differences between the test samples and controls were ascertained using the two sample t-test.
Results: For all of the test groups, Admira demonstrated minimal solubility compared to Ketac and Photac. However, after 24 h in an electric field of 27 V/m, Admira demonstrated the highest increase in solubility compared to the controls (3.47 vs control at 0.76 μmol/l). The second highest increase yielded the conventional glass ionomer cement Ketac (8.62 vs control at 5.28 μmol/l), and a minor increase in solubility showed the composite-based glass ionomer material Photac (38.73 vs control at 31.78 μmol/l) compared to the controls.
Conclusion: This study demonstrated that galvanic processes increase the solubility of glass ionomer and composite. Therefore, the time of storage, electric field strength, and contact of the material with the electrodes significantly influenced their solubility.

* Corresponding author. Department of Prosthetic Dentistry, Center and School of Oral Medicine, University of Cologne, 32 Kerpener Street, Cologne 50931, Germany.
** Corresponding author. Department of Prosthetic Dentistry, Center and School of Oral Medicine, University of Cologne, 32 Kerpener Street, Cologne 50931, Germany.
E-mail addresses: venuskatehsari@yahoo.de (V. Hassani Katehsari), wilh.niedermeier@uni-koeln.de (W. Niedermeier).

https://doi.org/10.1016/j.jds.2019.04.005

Introduction

Oral galvanism (OG) leads to mucosal sensitivities[1,2] such as battery taste in the mouth, xerostomia,[3] and oral and dental diseases such as pulpitis, leukoplakia, or lichen planus.[4] Medical symptoms such as back and shoulder pain and mental problems such as depression, fatigue, and insomnia[2] are also associated with OG. OG is triggered by the interaction of various dental alloys used for restorations and fillings, with saliva as an electrolyte.[5–7] Replacement of metal restorations and fillings with non-metal appliances often improves or even eliminates symptoms.[8,9]

New dental materials were introduced to dentistry as better alternatives to existing metallic appliances. Composite resins offer an alternative to amalgams that are commonly used for dental fillings. Composites and glass ionomer cements are used as core build-up materials. Composites consist of an organic dimethacrylate matrix, disperse inorganic fillers such as quartz, glass, ceramics, and silicon oxide, and a coated composite phase with silanes and copolymers.[10] Conventional glass ionomer cements mainly consist of polyacrylic acid and inorganic fillers that undergo an acid-base reaction and cure within 5–10 min of mixing.[11,12] In addition to conventional, metal-reinforced, and highly viscous glass ionomer cements, the plastic-modified form of glass ionomer cement was developed toward the end of the 1980s. The main component is methacrylic polyacrylic acid that can be photo-polymerized.[12]

The chemical properties of these compound materials demonstrate that galvanic elements are formed by different metallic materials in the oral cavity and have potentials of up to 1000 mV that can dissociate and reduce the stability of composites or glass ionomer cements used to restore dental abutments.[2] Therefore, the aim of this investigation was to assess whether OG or electric fields (EF) can impair the quality of dental core build-up materials.

Materials and methods

Specimens

Each of the 60 test specimens were produced from three fundamentally different material types to simulate the core build-up materials in the mouth: (1) Ketac Molar Universal Aplicap glass ionomer filling material; (2) Photac Fil Quick Aplicap light-curing glass ionomer filling material (both 3M-Espe, Seefeld, Germany); and (3) Admira,ormocer (Voco, Cuxhaven, Germany). Tables 1–3 show the composition and ingredients of the materials. All of the specimens were processed as specified by the manufacturer and molded using a silicone template. They were 1 mm thick and had an outer diameter of 10 mm and an inner diameter of 4 mm. The total surface of the specimens was 65.97 mm^2 (circle ring formula: $a = \pi*(10^2\text{-}4^2)/4 = 65{,}97\ mm^2$). The test specimens were ground with metal free diamond paper with a roughness of 800 (Telum K 800, Paul Sommer,

Table 1 Composition and information on ingredients of Ketac Molar Universal Aplicap.[13]

C.A.S. Registry Number	Ingredient	% by Relative Weight (Wt)
7732-18-5	Water	60–65
		[a]Trade Secret
29132-58-9	Copolymer of Acrylic Acid-Maleic Acid	30–40
		[a]Trade Secret
87-69-4	Tartaric Acid	5–10
		[a]Trade Secret

[a] The exact percentage/concentration and specific chemical identity of this composition has been withheld as a trade secret.

Table 2 Composition and information on ingredients of Photac Fil Quick Aplicap.[14]

C.A.S. Registry Number	Ingredient	% by Relative Weight (Wt)
29132-58-9	Polyethylene Polycarbonic Acid	30–50
868-77-9	2 Hydroxyethyl Methacrylate	25–50
7732-18-5	Water	20–30
72869-86-4	Diurethane Dimetharylate	3–5
Unknown	Magnesium Hema Ester	5–10

Table 3 Composition and information on ingredients of Admira.[15]

C.A.S. Registry Number	Ingredient	% by Relative Weight (Wt)
Unknown	Ormocer	10–25
1565-94-2	BIS-GMA	5–10
72869-86-4	Urethandimethacrylat	5–10
EINECS: 276-957-5		

Loebau, Germany) to achieve a uniform and even surface quality (Fig. 1). All of the test specimens were soaked in distilled water for 1 h to remove unlinked monomers and impurities.

Pre-tests

In preliminary tests, the materials were each stored in a glass container with 80 ml of sodium chloride (NaCl, 0.9%) solution for 24 h. Subsequently, 2 ml of sample solutions were obtained to conduct a metal screening test (multi-element analysis). Calcium and aluminum were superior as solubility parameters in the following experiments (Fig. 2).

Test procedure

To simulate oral conditions, the tests were conducted following a pseudo-realistic setup. 160 ml of 0.9% saline was used as electrolyte solution with a temperature of 26 °C and a pH of 6.8 ascertained at the beginning and end of each test using pH indicator strips (Neutralit pH 5.0—10.0, Merck, Darmstadt, Germany). The temperature of 26° (room temperature of the lab) was chosen because for technical reasons, the entire experimental setup could not be placed in an incubator. That is why the room temperature of the laboratory was used as environmental temperature. All conditions (temperature, humidity, air pressure etc.) were equal for every experiment. The test specimens were suspended in their central hole via a plastic isolated wire. 2 ml samples of the electrolyte were obtained after 1, 2, 3, and 24 h. The samples were pipetted after stirring the electrolyte solution. Each test was repeated three times under exactly the same conditions to obtain a mean value. To avoid dilution and falsifying the results, the glass container was not refilled with saline solution after each sample. For each process, the materials and tools used were exclusively made of glass or polyethylene. The experiments were divided into two series.

(1) For the first series, test specimens of the three dental materials Ketac, Photac, and Admira were suspended for 24 h in saline solution without an electric field (EF = 0 V/m).
(2) The second series was conducted in a similar sequence. The only difference was that two electrodes were added to the test setup and the samples were exposed to an electric field (Fig. 3). Fine gold sheet electrodes (99.9%, 2 × 3 inches) were attached using clamp stands and were aligned parallel to each other in the solution. The test specimens were angled at 90° to the electrodes connected with an auto-controlled voltage source. The EF was varied from 10 to 27 V/m by adjusting the sheet electrodes and modulating the voltage. The test material was exposed to the fields both with and without one-sided and two-sided contact with the electrodes. Thereby, direct contact with the electrodes should simulate the conditions at a dental abutment, where the metal crowns on one side and the root post on the other side are in direct contact with the core build-up material.

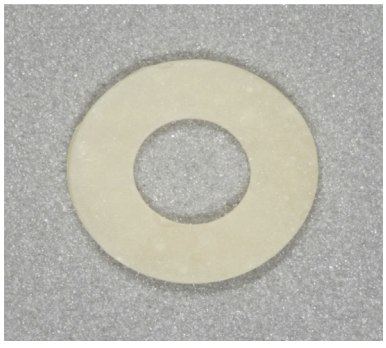

Figure 1 Photac Fil test specimen; the surface was ground flat using diamond paper (Telum K 800).

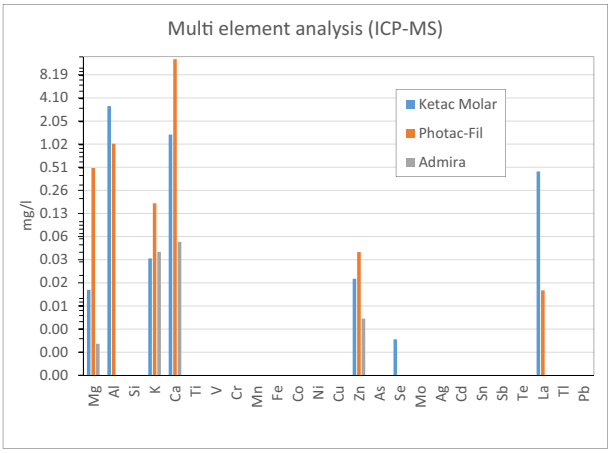

Figure 2 Detail components of the test materials by a Multi element analysis (ICP-MS). The elements Calcium and aluminum are superior solubility parameters which are contained most in the test subjects.

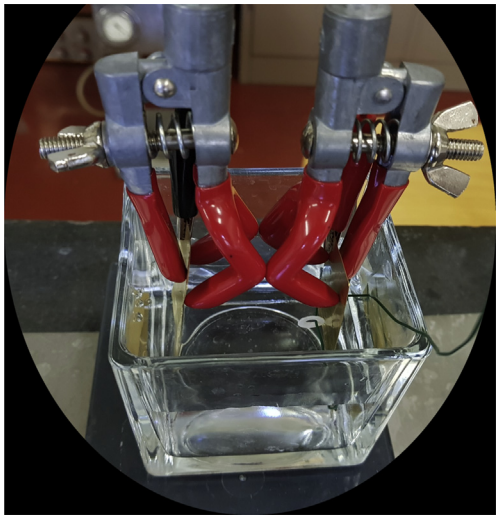

Figure 3 Specimen suspended with an insulated wire via a central hole in a glass containing 0.9% saline solution and in one-sided contact with one of two gold sheet electrodes (left and right).

Chemical and statistical analysis

The concentrations of the pipetted samples were analyzed by an accredited laboratory using mass spectroscopy with inductively coupled plasma (ICP-MS). Each sample was tested 3 times against the standard (electrolyte without sample).

The two sample t-test for independent samples was conducted to differentiate the test samples from the controls. The evaluations were carried out in a post hoc test according to Bonferroni's correction ($\alpha = 0.05$).

Results

Ketac

Table 4 demonstrates the solubility values of aluminum for Ketac Molar under one-sided contact (EF = 10 V/m, EF = 20 V/m) and under two-sided contact (EF = 10 V/m, EF = 27 V/m) of the electrodes vs the controls (EF = 0 V/m). After 3—24 h, all of the samples demonstrated an increase in their aluminum solubility. The differences in the controls were significant without exception for an EF of 20 and 27 V/m.

Photac

Table 5 shows the release of Ca from Photac under one-sided and two-sided contact with the electrodes against the controls. The values indicate an increase in the calcium concentrations for all of the field strengths and periods. After only two hours, the calcium solubility of all of the test samples had significantly increased compared to the controls.

Admira

Amira demonstrated an increase in calcium solubility after 2—24 h (Table 6). The increase was significant when the material had contact with both electrodes or at a field strength of 20 V/m or more.

Comparison of all materials

Fig. 4 demonstrates a comparison of the Ketac, Photac, and Admira materials in an EF of 27 V/m. The aluminum release was determined for Ketac and the calcium release was ascertained for Photac and Admira. Admira, although showing the maximum field- and time-dependent increase in solubility, demonstrated a minimum overall release of calcium. The maximum release of inorganic minerals was ascertained for Photac.

Discussion

The present study investigated the phenomenon of oral galvanism and whether electric fields can exert an influence

Table 4 Measurements of the Al concentration for Ketac Molar with one and two electrode contact. The mean values and their standard deviations are listed. The arrows indicate an increase (↑) or decrease in the concentrations vs the controls (0 V/m). The statistical significance of the differences (two sample t-test): *$p < 0.05$: noticeable increase and **$p < 0.01$: significant increase.

Time		1 h		2 h		3 h		24 h	
Field strength		MW µmol/l	SD µmol/l	MW µmol/l	SD µmol/l	MW µmol/l	SD µmol/l	MW µmol/l	SD µmol/l
0 V/m		**1.22**	0.19	**1.68**	0.43	**2.2**	1.12	**5.28**	1.78
10 V/m	One-sided contact	**1.25**	0.65	**1.70**	0.93	**2.62** ↑	0.92	**6.1** ↑	0.79
20 V/m	One-sided contact	**2.10** ↑*	0.54	**3.78** ↑**	0.63	**5.27** ↑**	1.04	**8.27** ↑*	1.27
10 V/m	Two-sided contact	**1.25**	0.8	**2.23** ↑	0.19	**3.35** ↑	0.76	**7.65** ↑*	1.94
27 V/m	Two-sided contact	**1.57** ↑	1.01	**2.79** ↑	0.98	**4.17** ↑*	1.48	**8.62** ↑**	0.69

Bold signifies the importance of the solubility of the products in statistics.

Table 5 Measurements of the Ca concentration for Photac with one and two electrode contact. The mean values and their standard deviations are listed. The arrows indicate an increase (↑) or decrease in the concentrations vs the controls (0 V/m). The statistical significance of the differences (two sample t-test): *$p < 0.05$: noticeable increase and **$p < 0.01$: significant increase.

Time		1 h		2 h		3 h		24 h	
Field strength		MW µmol/l	SD µmol/l	MW µmol/l	SD µmol/l	MW µmol/l	SD µmol/l	MW µmol/l	SD µmol/l
0 V/m		**1.99**	1.11	**3.92**	0.64	**5.9**	1.47	**31.78**	8.98
10 V/m	One-sided contact	**3.48** ↑*	1.15	**4.73** ↑*	1.29	**6.08** ↑	1.66	**33.63** ↑	0.81
20 V/m	One-sided contact	**4.95** ↑	4.02	**8.30** ↑*	3.46	**12.00** ↑**	4.64	**38.36** ↑	6.59
10 V/m	Two-sided contact	**3.63** ↑	0.48	**5.74** ↑*	0.17	**6.61** ↑	0.4	**34.37** ↑	0.54
27 V/m	Two-sided contact	**4.4** ↑*	2.15	**6.4** ↑*	0.9	**10.01** ↑**	2.9	**38.73** ↑*	2.32

Bold signifies the importance of the solubility of the products in statistics.

Table 6 Measurements of the Ca concentration for Admira with one and two electrode contact. The mean values and their standard deviations are listed. The arrows indicate an increase (↑) or decrease in the concentrations vs the controls (0 V/m). The statistical significance of the differences (two sample t-test): *$p < 0.05$: noticeable increase and **$p < 0.01$: significant increase.

Time		1 h		2 h		3 h		24 h	
Field strength		MW μmol/l	SD μmol/l	MW μmol/l	SD μmol/l	MW μmol/l	SD μmol/l	MW μmol/l	SD μmol/l
0 V/m		**0.34**	0.12	**0.4**	0.11	**0.49**	0.1	**0.76**	0.2
10 V/m	One-sided contact	**0.33**	0.09	**0.62** ↑	0.21	**0.78** ↑	0.39	**0.98** ↑	0.29
20 V/m	One-sided contact	**1.27** ↑*	0.35	**1.56** ↑*	0.5	**1.76** ↑*	0.59	**2.54** ↑*	0.84
10 V/m	Two-sided contact	**0.83** ↑*	0.2	**0.88** ↑*	0.17	**1.12** ↑*	0.24	**1.42** ↑*	0.3
27 V/m	Two-sided contact	**1.59** ↑*	0.5	**1.97** ↑*	0.69	**2.14** ↑*	0.64	**3.47** ↑*	1.29

Bold signifies the importance of the solubility of the products in statistics.

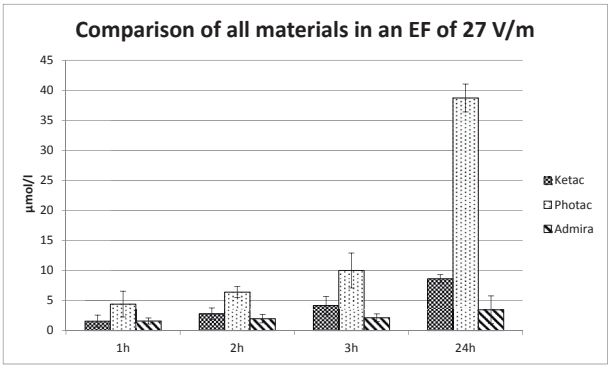

Figure 4 Comparison of the release of calcium and aluminum for Admira, Ketac, and Photac in an EF of 27 V/m under two-sided contact with the electrodes and their standard deviation. Since Ketac does contain less calcium, aluminum was used as a marker for this material. Therefore, the ordinate shows concentrations of aluminum (for Ketac) and calcium (for Admira and Photac) in μmol/l.

on dental core build-up materials. Whether the distance of the electrodes to the respective restorations is relevant and decisive for their solubility behavior must also be verified. In the mouth, different metallic restorations act as electrodes and saliva and tissue liquor serve as electrolytes.[16,17] Compared to non-precious metals, gold has a high standard potential and a very low tendency to corrode.[16] The corrosion processes was minimized using fine gold electrodes. A homogenous electric field was generated to a large extent in vitro by a low-frequency alternating voltage (1 Hz). The function of the voltage transformer was to prevent a sudden voltage drop between the gold electrodes and guarantee a constant current flow to minimize the polarization of the gold electrodes. It was set at a constant voltage between 0.1 V and 0.7 V to achieve electric field strengths between 10 and 27 V/m, corresponding to previous measured values found in vivo studies.[16–18] The electrolyte was 0.9% NaCl solution. With a pH value of 6.8, it fulfilled the ionic requirements of an electrolyte and also was very close to the pH value of saliva.[19,20] The glass container in our experiments was not refilled with saline solution after each sample. Otherwise the concentration of ions in the glass container would drop through the dilution and therefore retard the electrolysis of the test materials. The pretest showed which components have the highest solubility in Ketac Molar, Photac and Amira (Fig. 2).

Ketac Molar is a conventional glass ionomer cement. In addition to glass, oxide, and chemicals, it is mainly composed of polyacrylic acid that cross-links to form a calcium polycarboxylate gel.[11,12] Preliminary tests found that aluminum that was utilized to stabilize the material[12] demonstrated the highest solubility among all of the ingredients. Crisp, Lewis, and Wilson,[21] Sidler and Strub,[22] Mitchem and Gronas,[23] and Nathanson[24] studied the solubility of cements, including glass ionomer cements, and proved that they dissolved under certain conditions. Peez[25] and Frank demonstrated that Ketac Molar Easymix has a higher solubility in a neutral medium than in acid. The current study found that the solubility of aluminum increases in an almost neutral medium without EF; after exposure to an EF, a strong increase in the aluminum solubility occurs. When the test specimen came into contact with both electrodes, the aluminum solubility increased much more compared to the other experiments. Transferred to the oral cavity, this happens when the build-up material lies in between the crown and an endodontic metal post or pin. Depending on the electric potential generated by different metallic materials, in a crown-post situation, the EF can increase to 100 V/m and more. Moreover, the stability of nearby dentin and lining cements will also decrease as the result of a clinical failure.[18,26] In the present study, however, the EF had to be limited to 27 V/m for technical reasons to maintain the stability of the electrodes.[18,26] Therefore, it can be assumed that electrochemical dissociation is higher in vivo.

The solubility of the glass ionomer cement Photac Fil that was methacrylate polyacrylic acid was studied using calcium as a marker that demonstrated the highest solubility in the pre-tests.[27] Zankuli[28] observed the solubility of a plastic-modified Fuji II glass ionomer cement in distilled water. Without an EF, a rapid time-dependent increase in the Ca solubility was found in the 0.9% saline solution (pH = 6.8) used in the present study. Under one- and two-sided contact with the electrode, the concentration increased substantially. This means that Photac as a core build-up material would show a strong solubility behavior in the long run both without and with crowning.

Admira is an ormocer that is characterized by very good cross-linking properties and low monomer release.[29] Researchers such as Inoue and Hayashi investigated whether or not conventional composites can dissolve. They found components such as BIS-GMA and other residual monomers including camphorquinone in the eluates.[30,31] Admira demonstrated a very low solubility behavior in the current experiments. After 24 h without an EF, the calcium solubility in the medium doubled to 0.76 μmol/l compared to the beginning of the experiment, but the increase was not significant. However, as soon as electric potentials were applied via the gold electrodes, a significant increase in the Ca concentrations was detected. This indicates that the EF is more important than time and it strongly influences the solubility behavior of Admira. This raises the question of whether the increased solubility poses a danger to the stability of the crown when crowning using Admira as core build-up material. However, the measured values are in such a minimal range that they are less relevant for a decisive loss of core stability. At an EF of 27 V/m, Admira demonstrated 3—4 times higher Ca solubility compared to the controls at the beginning. The comparison of the materials tested demonstrated that the light-curing glass ionomer cement Photac was the most soluble both with and without an EF. The second highest solubility by time and EF was exhibited by the conventional glass ionomer cement Ketac. In general, Admira proved to have minimal time- and EF-dependent solubility and as a result, presumably the smallest loss of stability.

In conclusion and within the limitations of this in vitro study, it can be stated that clinically relevant galvanic processes in the mouth also exert an influence on the solubility of dental core build-up materials. Therefore, the use different types of alloys should be avoided for reconstructive measures in the oral cavity to minimize the dissolution of core build-up materials, luting cement,[18] or hard tooth substances[26] apart from the medical consequences for patients.

Conflicts of interest

The authors have no conflicts of interest relevant to this article.

References

1. Hampf G, Ekholm A, Salo T, et al. Pain in oral galvanism. *Pain* 1987;29:301—11.
2. Hugoson A. Results obtained from patients referred for the investigation of complaints related to oral galvanism. *Swed Dent J* 1986;10:15—28.
3. Bujas Z. Electrical taste. *Handb Sensory Physiol* 1971;4:180—99.
4. Martin MD, Broughton S, Drangsholt M. Oral lichen planus and dental materials: a case-control study. *Contact Dermatitis* 2003;48:331—6.
5. Chase HS. Oral electricity. *Dent Cosmos* 1879;21:205—7.
6. Meiners H. Electrical phenomena associated with metal fillings. *ZWR* 1984;93(41—4):46—7 [Article in German].
7. Meyer RD, Meyer J, Taloumis LJ. Intraoral galvanic corrosion: literature review and case report. *J Prosthet Dent* 1993;69:141—3.
8. Kappert HF, Ulbrich J, Gläss P, Huber H, Neumüller H. Determining the threshold level for sensibility responses to galvanic potentials in the oral cavity. *Dtsch zahnärztl Z* 1989;44:50—1 [In German, English abstract].
9. Nilner K, Glantz PO, Ryge G, Sundberg H. Oral galvanic action after treatment with extensive metallic restorations. *Acta Odontol Scand* 1982;40:381—8.
10. Koplin C. *Entwicklung eines Materialmodells für zahnärztliche Füllungen auf Kompositbasis. Schriftenreihe Werkstoffwissenschaft und Werkstoffechnik*. thesis. School of Dentistry Karlsruhe, 2007 [Article in German].
11. Espe 3M. *Ketac molar Aplicap instructions for use, 44000768158/01*. Seefeld: 3M Espe, 2014.
12. Boer WM. *Metallfreie restaurationen*. 4nd ed., vol. 1. Balingen: Spitta Verlag, 1999 [In German].
13. Espe 3M. *Ketac molar Aplicap material safety data sheet*. Seefeld: 3M Espe, 2014.
14. Espe 3M. *Photac Fil Quick Aplicap material safety data sheet. Seefeld*. 3M Espe, 2010.
15. Voco. *Admira/caps safety data sheet*. Cuxhaven: Voco, 2008.
16. Grünenberg S. *Oraler Galvanismus und DMPS-Test., thesis*. Aachen: TH Aachen, 2006:4—27 [Article in German].
17. Baars G, Christen HR. *Chemie*, 1st ed. Bern: Bildungsverlag, 2008:329—30 [In German].
18. Novakovic D. *Löslichkeit dentaler Befestigungsmaterialien im elektrischen Feld*. MED thesis. Cologne: University of Cologne, 2012 [Article in German].
19. Mandel I. The function of salvia. *J Dent Res* 1987;66:623—7.
20. Kabi Fresenius. *Isotonic sodium chloride 0.9%, instructions for use, 068 8971/00*. Bad Homburg: Fresenius, 2012.
21. Crisp S, Lewis BG, Wilson AD. A study of erosion and water absorption in both neutral and acidic media. *J Dent* 1980;8:68—74.
22. Sidler P, Strub JR. In vivo study of the solubility and sealing capacity of 3 fixation cements. *DZZ* 1983;38(5):564—71 [Article in German].
23. Mitchem JC, Gronas DG. Continued evaluation of the clinical solubility of luting cements. *J Prosthet Dent* 1981;3:289—91.
24. Nathanson D. No conclusive evidence for caries-inhibitory effect of glass-ionomer restorations in vivo. *Evid Based Dent* 2000;2:99.
25. Peez R, Frank S. The physical—mechanical performance of the new KetacTM Molar Easymix compared to commercially available glass ionomer restoratives. *J Dent* 2006;34(8):582—7.
26. Kunze M. *Löslichkeit von Zahnhartsubstanzen im elektrischen Feld*. MED thesis. Cologne: University of Cologne, 2014 [Article in German].
27. Braden M, Clauston EE, Clarke RL. Diffusion of water in composite filling materials. *J Dent Res* 1976;55:730—2.
28. Zankuli MA, Devlin H, Silikas N. Water sorption and solubility of core build-up materials. *Dent Mater* 2014;30(12):324—9.
29. Wolter H. Werkstoffe mit Biss, Teil 1: ormocere, Würzburg, *DZW* Ausgabe 11/15. [Article in German]
30. Inoue K, Hayashi I. Residual monomer (Bis-GMA) of composite resins. *Oral Rehabil* 1982;9:493—7.
31. Spahl W, Budzikiewicz H, Geurtsen W. Study on the residual monomer contents of different light curing hybrid composite resins. *DZZ* 1991;46:471—5 [In German, English abstract].