Assessment of Physical and Chemical Parameters of Four Endodontic Sealants

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The endodontic sealants have an essential role in the root canal filling. The aim of this study was to analyse and compare three physio-chemical properties (pH, solubility, viscosity). The most significant decrease of the viscosity was for MTA-Fillapex, followed by Endoflas FS and Acroseal. The best behavior was recorded for Endomethasone N. The total decrease of dry mass (reported to baseline) was 0.0113 for Acroseal. Endoflas FS had a total decrease of dry mass of 0.0377 (from 0,6864 to 0.6487) during the study. The total decrease of dry mass (reported to baseline) was 0.0047 for Endomethasone N. MTA Fillapex had a total decrease of 0.0226 (from 0.4805 to 0.4579) during the study period. MTA Fillapex had the highest pH values (pH 10.16), followed by Acroseal (pH 8.79), Endoflas FS (pH 7.98) and Endomethasone N (pH 7.73).

Keywords: pH, solubility, viscosity, endodontic sealants.

The endodontic sealants have an essential role in the root canal filling. These materials fill the empty endodontic spaces left by the inability of guttapercha to fill and and adhere to dentinal wall (in cold lateral filling techniques). The sealing acts like a bonding between dentine and guttapercha core, as element that complete the cones sealing and as lubricating agent during the cone insertion (De-Deus G et al. 2015). No endodontic sealants can ensure an ermetic closure of the periapical area, but can initiate various degrees of short time periapical inflammations that do not seem to influence the healing processes *restitutio ad integrum* of the periapical tissues (Kaur A et al. 2015).

Experimental part

The aim of study

The aim of this study was to analyse and compare three physio-chemical properties *(p*H, solubility, viscosity) of four endodontic sealants: Acroseal, Endomethasone N, Endoflas FS, MTA-Fillapex.

Material and methods

The solubility, *p*H and viscosimetry were determined accordingly to the methods recommended by American National Institute of Standardisation and American Dental Association (ANSI /ADA, no.57), proposed also by some researchers (Duarte MA et al. 2000, Versiani MA et al. 2006, Gambarini G et al. 2006).

Vîscosity measurements

The rheological properties of endodontic sealants were measured using reometer Physica MCR 501. The materials were prepared as pastes and applied on reometer plaque. For an accurate comparative analysis, the samples were weighted using a digital balance (CP124S, Sartorius, Germania) at 37° C (with a 0.1mg reading. The viscosity was analysed using dinamic shearing tests. For the measurements of shearing forces, it was fixed a rate *Sher* - from 0.001 to 100 s^{-1}. The viscosity value was recorded as mean value of the measurements of 15 samples from each endodontic sealant (recorded at 37° C), and the mean value was recorded.

Solubility

The test measures the stability of endodontic sealants after the material setting. The samples were immersed in 20-50 mL deionized water for 24 h.The next sealing materials were studied: Acroseal (SEPTODONT, France); Endomethasone N (SEPTODONT, France); Endoflas FS (SANLOR, Colombia); MTA Fillapex (ANGELUS, Brasil).After the sample removal from water, the presence of residual materials was checked (indicating the solubility of the material), and both sample and residual materials were weighted to measure the quantity of solubilised material. The acceptable total mass loss limit was set at 3%. Fifteen samples (1.5 mm thickness, inner diameter 7.75 mm) were prepared and introduced in special matrices. The samples were weighted with a precision of 0.0001 g (ABT 220-4M, Kern & Sohn GmbH, Germany). Each sample was weighted three times and it was calculated the mean value of the three results. The samples were immersed in 15 mL deionized water and maintained for 24 h to 37 ± 2 Celsius degrees. After this time interval the water excess was removed with absorbant paper, its were preserved for 24 h in desumidificator and weighted for a second time. The solubility was calculated as mass percentual loss in relation to initial mass. The procedure was repeated three times for each endodontic sealant.

*p*H measurements

This test measures *p*H of the endodontic sealings. The technique was applied after a time interval three times higher than the the setting time of the material. The samples were immersed in recipients with 30 mm diameter and filled with 50 mL bidistilled water (tip Milli-Q) with neutral *p*H. Fifteen samples were prepared for each material. The samples were sealed for 24 h in a recipient with 15 mL water double-distilled (ddH2O) at 37°C. After 24 h it was performed the first *p*H recording.The testing of double-distilled water was performed using pH-meter - (InoLab® pH 720 Laboratory pH Meter/Germany) at a time interval of 24h, 48h, 7, 14 and 21 days after the products mixing. The measurements for each sample were performed three times (calibration of *p*H was performed

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using a solution with a *p*H range 3-7). The mean values and the standard deviations were calculated for all measurements.

To investigate if the differences between the behavior of the four endodontic sealants for 6 time moments were statistically significant, we used non-parametric test Kruskal-Wallis; the test post-hoc Tamhane was used to localise the differences between the four materials.

Results and discussions

The investigated endodontic cements had a viscosity decreased directly related to the increase of shearing speed (shearing behavior to thinning) at temperature 37°C (fig. 1). The analysis of the behavior of these cements was similar to that of the pseudo-plastic concentrated suspensions. The most significant decrease of the viscosity was for MTA-Fillapex, followed by Endoflas FS and Acroseal. The best behavior was recorded for Endomethasone N.

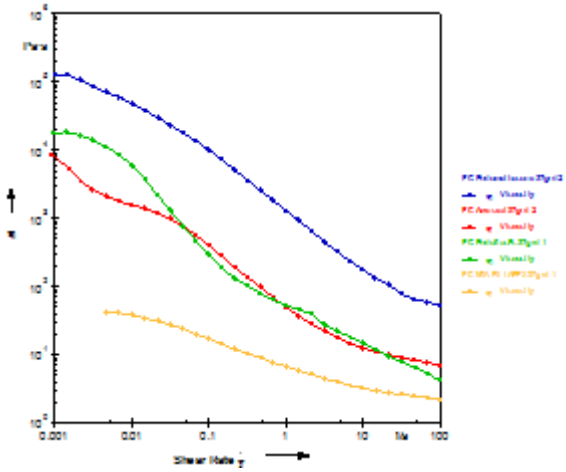

Fig. 1. Shearing behavior to thinning for the investigated sealants

Table 1 describes the statistical evolution of the solubility (dry mass) for all the analysed endodontic sealants.

The comparative analysis of the dry mass and the mass differences of the investigated sealantsrelated to the baseline highlight the following results (figs. 2,3).

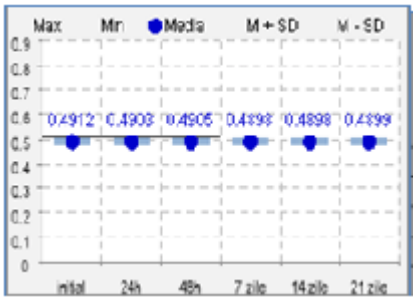

Fig. 2. Mean value of dry mass for Acroseal

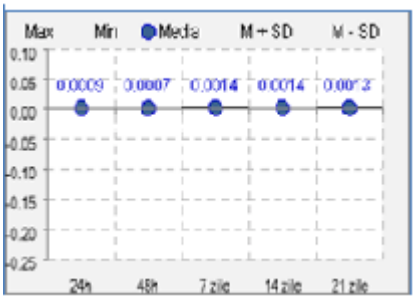

Fig. 3. Mean values of mass difference for Acroseal

The solubility of Acroseal decreased after 24h (from 0.4912 to 0.4903) followed by an increase of 0.0002 after 48h. After 7 days the solubility decreased with 0.0107 (comparing to the value recorded at 48h), and has maintained unchanged until the day 21, when it was stabilized to 0.4899. The total decrease of dry mass (reported to baseline) was 0.0113 (figs. 2,3).

Sealant		BASELINE_ Mass	24h_ Mass	48h_ Mass	7days_ Mass	14days_ Mass	21days_ Mass
Acroseal	N	12	12	12	12	12	12
	Mean	.491200	.490267	.490467	.489800	.489833	.489867
	Mean std. error	.0044649	.0043363	.0043334	.0042300	.0041971	.0041927
	std. deviation	.0154668	.0150213	.0150113	.0146531	.0145391	.0145238
	Minimum	.4769	.4768	.4767	.4765	.4767	.4767
	Maximum	.5116	.5102	.5103	.5092	.5091	.5091
Endoflas FS	N	12	12	12	12	12	12
	Mean	.686367	.678433	.670533	.665333	.657200	.648667
	Mean std. error	.0088651	.0086222	.0086064	.0085804	.0085095	.0084811
	std. deviation	.0307095	.0298681	.0298135	.0297232	.0294778	.0293796
	Minimum	.6450	.6382	.6303	.6252	.6174	.6090
	Maximum	.7107	.7021	.6935	.6880	.6797	.6711
Endomethasone	N	12	12	12	12	12	12
	Mean	.523633	.521367	.517433	.520267	.519567	.518967
	Mean std. error	.0114793	.0118888	.0109443	.0118298	.0118917	.0119764
	std. deviation	.0397656	.0411840	.0379123	.0409795	.0411942	.0414875
	Minimum	.4703	.4660	.4661	.4652	.4642	.4632
	Maximum	.5567	.5548	.5432	.5537	.5531	.5527
MTA Fillapex	N	12	12	12	12	12	12
	Mean	.480533	.475233	.469373	.466733	.462067	.457933
	Mean std. error	.0041343	.0040259	.0039557	.0039707	.0039705	.0040815
	std. deviation	.0143216	.0139460	.0137029	.0137548	.0137543	.0141386
	Minimum	.4619	.4572	.4520	.4494	.4451	.4405
	Maximum	.4945	.4891	.4837	.4813	.4772	.4735

Table 1
THE EVOLUTION IN TIME OF THE DRY MASS FOR THE INVESTIGATED SEALANTS FROM BASELINE TO DAY 21. MEAN, STANDARD DEVIATION

Endoflas FS had a total decrease of dry mass of 0.0377 (from 0.6864 to 0.6487) during the study, with a constant decrease tendency (0.008 between the investigated moments); the only time period recorded with a lower decrease was from 48h to 7 days (mass variation only 0.0052(figs. 4, 5).

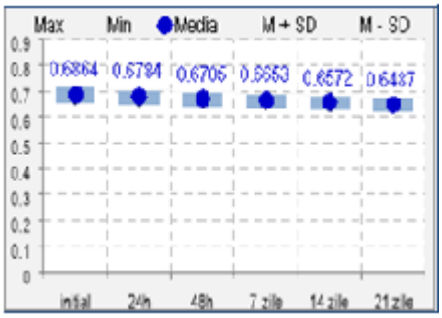

Fig. 4. Mean value of dry mass for Endoflas FS.

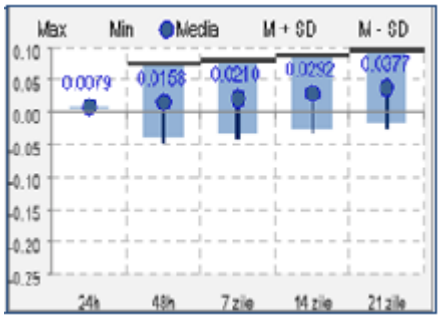

Fig. 5. Mean value of mass difference for Endoflas FS

The solubility of Endomethasone N decreased with 0.0022 after 24h (from 0.5236 to 0.5214). After 48h it was recorded another decrease of 0,004 (comparing to 24h dry mass), followed by an increase of 0.0029 after 7 days (comparing to 48h dry mass). After 21 days the solubility decreased with 0.0006 (comparing to the value recorded at 14 days). The total decrease of dry mass (reported to baseline) was 0.0047 (figs. 6,7).

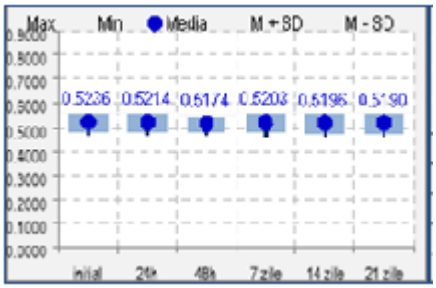

Fig. 6. Mean value of dry mass for Endomethasone N

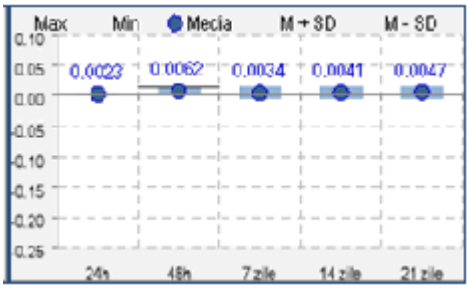

Fig. 7.Mean value of mass difference forEndomethasone N

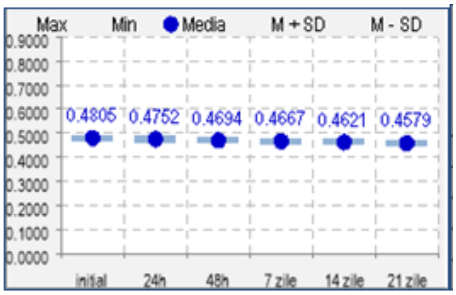

Fig. 8. Mean value of dry mass for MTA Fillapex

MTA Fillapex had a total decrease of 0.0226 (from 0.4805 to 0.4579) during the study period; the dry mass loss was (between the investigated moments) 0.005 (excepting the variation from 48h to 7 days, with only 0.0027) (fig. 8, 9).

Sealant		24h_ Mass_diff	48h_ Mass_diff	7days_ Mass_diff	14days_ Mass_diff	21days_ Mass_diff
Acroseal	N	12	12	12	12	12
	Mean	.000933	.000733	.001400	.001367	.001333
	Mean std. error	.0001781	.0001356	.0002462	.0002832	.0002832
	std.deviation	.0006169	.0004697	.0008528	.0009810	.0009810
	Minimum	.0001	.0002	.0004	.0002	.0002
	Maximum	.0014	.0013	.0024	.0025	.0025
Endoflas FS	N	12	12	12	12	12
	Mean	.007933	.015833	.021033	.029167	.037700
	Mean std. error	.0002429	.0158666	.0158175	.0157558	.0157312
	Deviatia std.	.0008414	.0549634	.0547935	.0545797	.0544943
	Minimum	.0068	-.0485	-.0430	-.0347	-.0261
	Maximum	.0086	.0804	.0855	.0933	.1017
Endomethasone	N	12	12	12	12	12
	Mean	.002267	.006200	.003367	.004067	.004667
	Mean std.error	.0004621	.0016560	.0030505	.0030467	.0030600
	std.deviation	.0016008	.0057366	.0105673	.0105542	.0106003
	Minimum	.0006	.0007	-.0098	-.0092	-.0088
	Maximum	.0043	.0137	.0148	.0153	.0157
MTA Fillapex	N	12	12	12	12	12
	Mean	.005300	.011160	.013800	.018467	.022600
	Mean std. error	.0001371	.0003627	.0004141	.0006072	.0005990
	std.deviation	.0004748	.0012565	.0014346	.0021034	.0020750
	Minimum	.0047	.0099	.0125	.0168	.0210
	Maximum	.0058	.0128	.0157	.0213	.0254

Table 2
THE EVOLUTION OF THE SEALANTS MASS DIFFERENCE FROM BASELINE TO DAY 21. MEAN, STANDARD DEVIATION.

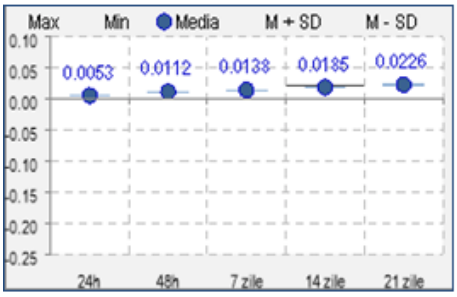

Fig. 9. Mean value of dry mass for MTA Fillapex

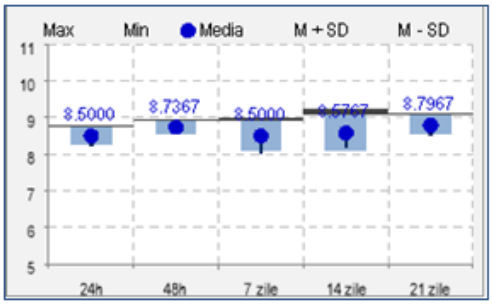

Fig. 10. Mean values of pH forAcroseal.

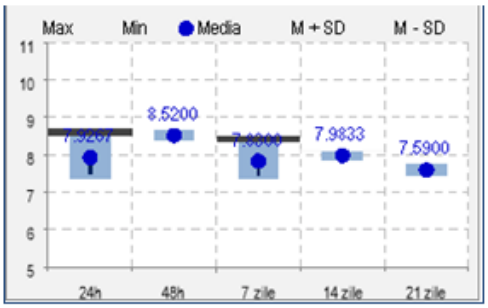

Fig. 11. Mean values of *p*H for Endoflas FS

The table 2 presents, from statistic perspective, the evolution of *p*H for all the investigated endodontic sealants (table 2).

Acroseal *p*H was 8.50 at 24h, and increased to 8.73 at 48 h. After 7 days *p*H returned to 8.50, and increased to 8.57 at day 14, and to 8.79 at day 21 (fig. 10).

Endoflas FS *p*H was 7.92 at 24h, and increased to8.52at 48 h. After 7 days *p*H decreased to7.83, and had a slight increase to 7.98 at day 14, and to 7.59 at day 21 (fig. 11).

Endomethasone N *p*H was 7.23 at 24h, and increased to 7.73 at 48 h. After 7 days *p*H returned to 7.02, and had a slight increase to 7.43 at day 14, followed by a decrease to 7.10 at day 21 (fig. 12).

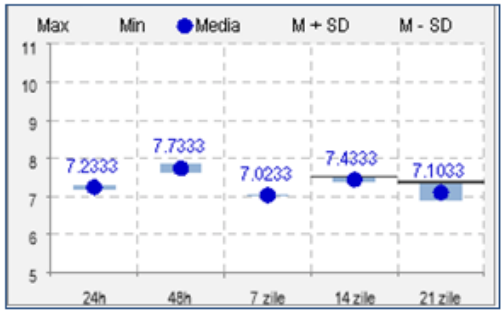

Fig. 12. Mean values of *p*H forEndomethasone N.

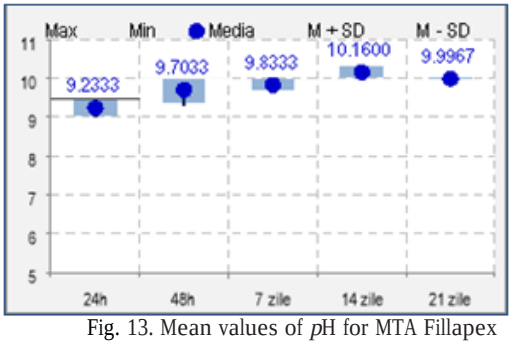

Fig. 13. Mean values of *p*H for MTA Fillapex

MTA Fillapex *p*H was 9.23 at 24h, and increased to 9.70at 48 h. After 7 days *p*H increased to 9.83, and overpassed 10.16 at day 14, followed by a slight decrease to 9.99 at day 21 (fig. 13).

The four investigated endodontic selalants had a significant statistical different behavior (related to the evolution of *p*H values) at the five time moments during the study. Tamhane test for multiple comparisons showed that pH differences between sealants were significant statistical, for all five tested moments.

Many researchers aimed to compare biological, physical and chemical properties of various endodontic sealants (Rai RU et al. 2016, Akcay M et al. 2016, Marciano MA et al. 2016). This research analysed the solubility, *p*H, viscosimetry ofsome frequently used endodontic sealants in dental practice (Endomethasone N®, Acroseal®, Endoflas FS®), and the results were compared with values recorded for MTA-FillApex®. The endodontic sealants must posess a low solubility level when in contact with periapical fluids. If the endodontic sealants would have high solubility, the chemical compounds would be fast released and would irritate the apex, initiating local inflammation. When voids and spaces are formed between root canals walls and

Table 3

DIFFERENCES BETWEEN *p*H VALUES FOR THE INVESTIGATED ENDODONTIC SEALANTS

Kruskal – Wallis test for de rank comparisons	*Acroseal*	*Endoflas FS*	*Endo metahasone N*	*MTA Fillapex*	Value Chi squared
	Mean rank	Mean rank	Mean rank	Mean rank	**Sign. *p***
Analysis at 24h	42.50	27.83	21.17	6.50	41.448
					.000
Analysis at 48h	42.50	29.17	19.83	6.50	42.618
					.000
Analysis at 7 days	42.50	28.50	20.50	6.50	42.053
					.000
Analysis at 14 days	42.50	30.50	18.50	6.50	44.371
					.000
Analysis at 21 days	42.50	30.50	17.83	7.17	43.592
					.000

guttapercha mass, are created favourable conditions for the bacterial multiplication and growth (Rai RU et al. 2016, Akcay M et al. 2016, Marciano MA et al. 2016). The results of actual study can be discussed and compared with literature data of related to in vitro researches (pH-meters, spectrophotometry) that assess *p*H values and buffering ability by the recording of calcium ions levels released at various time intervals (Kuga MC et al.2014, Canadas et al.2014, Zhou HM et al.2013).

Kuga MC et al. (2014) analysed the thinning level, *p*H values and calcium ions release rate for MTA Fillapex (MTAF) with 5% (MTAF5) or 10% (MTAF10) calcium hydroxide (HC), values that were compared with AH Plus. After 24 hours, 7 and 14 days, *p*H was determined for each sample. Regarding values after 24 h: MTAF5 = MTAF10 = MTAF > AH Plus; after 7 and 14 days: MTAF5 = MTAF10 > MTAF > AH Plus. The addition of 5% calcium hydroxide to MTA Fillapex (weight) is an alterantive to reduce the high level of thinning for this sealants, without altering its alkalinisation ability in time. Zhou HM et al. (2013) assessed *p*H, viscosity and other physical properties of MTA Fillapexand Endosequence BC. *p*H values were compared with two epoxidic resins- AH Plus and ThermaSeal, with a silicone-based sealant- Gutta-Flow- and a zinc-oxyde-eugenol sealant- Pulp Canal Sealer. The *p*H changes were recorded at 24 h after mixing and at 5 weeks after final setting. The viscosity was investigated at different intervals (72, 10 and 5 mm/min) laat room temperature, using Instron 3360 testing system. ISO 6876/2001 recommendations were respected regarding thinning, volumetric change, solubility and thickness. The sealant MTA Fillapex had higher thinning than a prezentat Endosequence BC ($p < 0.05$). MTA Fillapex and Endosequence BC had highest thickness of the sealing paste. Endosequence BC had the highest value of solubility, accordingly to maximum mass fraction 3% recommended by ISO 6876/2001, as well as acceptable dimensional change. Regarding *p*H, the investigated sampleshad alkaline values for all the investigated moments. The *p*H values for AH Plus and ThermaSeal were alkaline at baseline and decreased significantly after 24 hours. The viscosity of the tested sealants increased while the injection volume decreased. Grga D et al. (2011) compared the weight changes ofAcroseal, Apexit, calcium hydroxide sealants, and AH Plus sigilant for different exposure times. The results of the study confirmed the importance of low solubility as the most important physical property of an endodontic sealant. The therapeutic effect of calcium hydroxide endodontic cement depends on ions level, related to the material partial solubility. The samples were weighted and immersed in Hank solution for 1 h, 24 h, 96 h, 14 days and 28 days. After the exposure periode, the samples were removed from the deionised water, dried and weighted again. The mean weight changes were determined and statistically analysed. The highest mass differences were observed for Apexit (1.52%) and were significant different fromAcroseal (0.93%) ($p < 0.05$) and AH Plus (0.45%) ($p < 0.05$). The differences were not statistically significant between Acroseal and AH Plus (excepting the recording after 96 h). The conclusion was that Acroseal had a behavior more similar to AH Plus, and more different from Apexit. Poggio C et al. (2010) tested the solubility of six different sealants, two sealants containing zinc-oxide-eugenol (Endomethasone C, Argoseal), two sealants calcium-hydroxide-based (Bioseal Normal, Acroseal) and two sealants with resins (AH Plus, MM Seal). The solubility was determined by measuringthe weight loss of samples (%)after 24 h and after 2 months. The test was performed accordingly to ISO 6876 , as well as ADA (no.30, 57). The data were statistically analysed using ANOVA one-way test for significant statistical differences between groups. Post-hoc test showed that resin-based sealants had lower solubility ($p< 0,05$) comparing with the other investigated sealants, without significant statistical differences ($p> 0,05$). The weight loss was less than 3%. In our study, all sealants had low solubility, and the lowest solubility values were recorded for resin-based sealants that can be considered, due to this property, close to the ideal endodontic sealant.

Conclusions

Endoflas FS had the highest value for dry mass (mean value 0.6864) and MTA Fillapex had the lowest value for dry mass (mean value 0.4805), close to Acroseal (mean value 0.4912). Endomethasone N is in an intermediate position, with mean dry mass value 0.5236. MTA Fillapex had the highest pH values, followed by Acroseal, Endoflas FS and Endomethasone N. The investigated endodontic cements had a decrease of viscosity related to the increase of shearing speed, at temperature 37°C. The most significant decrease of viscosity was recorded for MTA-Fillapex, followed by Endoflas FS and Acroseal, while Endomethasone N had the highest viscosity values.

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