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Particle size effects on abrasion, surface polishing and stain removal efficacy in a tooth model system

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2	Particle Size Effects on Abrasion, Surface Polishing and Stain
3	Removal Efficacy in a Tooth Model System
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7	Short title: Toothpaste Abrasive Particle Size Effects
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36 Highlights

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The existence of a critical particle size (CPS) for toothpaste abrasive has been
demonstrated for the first time in a tooth model system.

A linear relationship was observed between cleaning power and enamel abrasion.

• A linear association was shown between enamel abrasion and the number of

toothbrush strokes.

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43 Abstract

Four calcined alumina abrasive particles [ultrafine (0.05 µm), 3 µm, 9 µm and 20 µm] with 44 45 defined sizes were investigated for their effects on toothbrush abrasion, surface polishing and 46 stain removal in vitro. The existence of a critical particle size (CPS) was shown for the first time in a tooth model system and in the present study a CPS of ~2.3 µm for d10, 4.3 µm for 47 48 d50 or 7.8 µm for d90 for the calcined alumina abrasives was apparent. The d10, d50 and d90 values indicate that 10%, 50% and 90% of the particles measured were less than or equal to 49 the size stated. This dimension enabled maximum abrasive action on the tested specimens 50 resulting in the largest wear depth, greatest surface polishing and best stain removal. The 51 enamel wear depth decreased when brushed with abrasives above the critical particle size and 52 53 became almost independent of further particle size increases, which is useful for minimising wear effects in the development of dentifrice. The findings provide new information on 54 abrasive particle size for modification and control of toothpaste abrasivity and cleaning. 55 56

57 Keywords

Bovine enamel; Toothbrush abrasion; Surface polishing; Stain removal; Critical particle size

61 1. Introduction

Oral health is essential to general health and quality of life. Toothbrushing with toothpaste 62 aims to remove the dental plaque biofilm, food debris and dental stain on accessible tooth 63 64 surfaces [1]. Indeed, there is an increasing demand for cleaning products which improve dental aesthetics [2]. The potential adverse effect, however, of toothbrushing with toothpaste 65 is tooth abrasion [3,4] and this can lead to dentine hypersensitivity, dental pain, and poor 66 67 aesthetics [5,6]. Ideally, a toothbrushing regime should show excellent cleaning efficiency for dental plaque and stain removal, as well as for polishing ability, while exerting minimal tooth 68 wear [7-9]. It is therefore important that we develop new toothpaste formulations and 69 toothbrush designs which optimally "clean" whilst minimising dental hard tissue damage. 70 71 72 Dentifrices or toothpastes are formulated with dental abrasive particles [10] which are the key

⁷² Definitives of toothpastes are formulated with definal abrasive particles [10] which are the key
⁷³ components that physically clean and polish the tooth surfaces [11,12]. Historically, the
⁷⁴ commonly used dental abrasives included silica, alumina, dicalcium phosphate, calcium
⁷⁵ carbonate, sodium carbonate, hydroxyapatite, perlite and diamond [8,13-17]. A significant
⁷⁶ number of studies have investigated the factors contributing to good oral hygiene, including
⁷⁷ stain removal and tooth wear. Data indicate that properties of the abrasive particles, including
⁷⁸ their hardness, shape, size, size distribution and concentration are key to cleaning
⁷⁹ performance [7,9,10,12,16-18].

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The amount of toothpaste abrasives added in the toothpaste formulation varies with the abrasive properties used. The most commonly used abrasives are hydrated silica and calcium carbonate, and they are included in the range of 8 w/w% to 20 w/w%; alumina and perlite, however, are included at lower concentrations of 1 w/w% to 2 w/w%, due to their higher

enamel abrasivity [16]. Furthermore, particle sizes and their size distributions are important 85 factors in determining abrasivity and stain removal [12]. Generally the use of abrasives with 86 87 relatively broad particle size ranges in dentifrices has previously precluded an understanding of the influence of particle size. However, the size effect is a well-known phenomenon in 88 abrasion and it is understood that there is a minimum abrasive particle size or critical particle 89 size (CPS) which allows maximum abrasive action [16,19,20]. Reportedly, wear or wear rate 90 91 increases as abrasive particle size also increases up to a threshold or a CPS, after which the wear rate becomes almost independent of further particle size increases. This CPS is 92 93 frequently reported as being approximately 100 µm [20], however this value is variable dependent upon the different substrates and abrasives used, e.g. 45 µm for copper on copper, 94 and 90 µm for nylon on nylon [19]. Indeed, a particle size of ~40 µm is reported as optimal 95 when metals, such as Silver Ag, Cadmium Cd and Molybdenum Mo or dental tissues 96 97 (including enamel and dentine) abraded against SiC grits with a diameter range of 13-125 µm [1]. 98

99

In a tooth model system, limited knowledge exists with regard to the influence of the abrasive 100 particle size and size distributions on tooth wear and stain removal [16]. Data indicate, 101 however, that toothpastes with larger particle sizes produce more wear than those with 102 smaller particles [21-23, 24], although results in one recent report suggested no effect of the 103 104 particle size on enamel erosion and abrasion [25]. Similarly there was a lack of association between average particle size and enamel wear when three sizes of commercially produced 105 silicon carbide grit with a diameter range of 14-73 µm were used as abrasive particles [26]. 106 107 Interestingly, a linear relationship has been found between abrasive particle size and enamel abrasion when enamel was brushed with bioactive glass containing toothpastes [23]. Larger 108 particles also showed higher dentine abrasion effects when brushing was performed using 109

calcium carbonate (two particle sizes with an average particle diameter of 7 µm and 15 µm, 110 respectively) and aluminium hydroxide (two particle sizes with an average particle diameter 111 of 8 µm and 13 µm, respectively). No impact of particle size on enamel erosion/abrasion was 112 found when eroded enamel was brushed with commercial toothpastes consisted of abrasive 113 particles above 40 µm [25]. 114

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116 To our knowledge there have been no studies which have explored the existence of a CPS in a tooth model system. Consequently, we hypothesised that there would exist a CPS in a tooth 117 118 model system. Therefore, the aim of this study was to investigate the effect of calcined alumina abrasives with defined particle size and size distribution on toothbrush abrasion, 119 polishing and stain removal. 120

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2. Materials and methods

2.1. Characterisation of calcined alumina abrasives 123

124 Four calcined alumina abrasives (Almatis GmbH, Ludwigshafen, Germany) [designated as 125 ultrafine (0.05 μ m), 3 μ m, 9 μ m and 20 μ m] with defined particle size were used in the present study. Particle size analysis was performed on a Malvern Mastersizer 2000 laser 126 diffraction particle size analyser (Malvern Instruments Ltd, United Kingdom). Calcined 127 128 alumina abrasives were added in distilled water to an agitated flask attached to the diffraction machine and the particle size distribution of the abrasives was determined. Particle 129 distribution graphs are shown as average values of three measurements, error bars were 130 negligible and were omitted. 131

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133 The morphology of the calcined alumina abrasive particles was observed under Scanning Electron Microscopy (SEM, EVO MA10, Zeiss). The alumina abrasive particles were 134

adhered to carbon tape on an aluminium stub (Agar Scientific Ltd., UK). Ultra-thin sputtered
conductive coating (gold) (EMITECH K550X, Emitech, United Kingdom) was applied to the
particles prior to SEM observations to prevent charging of the specimens. Representative
images were captured under a range of magnifications.

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2.2 Preparation of bovine enamel samples

141 Bovine permanent incisor teeth were collected and stored in 0.1% (w/w) thymol (Sigma-Aldrich, UK) solution at 4 °C prior to use. Tooth crowns were obtained from the dissection of 142 143 bovine teeth using a custom-built diamond-edged saw with water cooling. Bovine enamel specimens (approximate $12mm \times 18mm$) were prepared following embedding tooth crowns 144 in Ø25 mm blocks of epoxy resins (Buehler, UK). A Phoenix Beta Grinder/Polisher (Buehler, 145 UK) was used with SiC abrasive discs (Buehler, UK) for sample preparation. Eight bovine 146 enamel specimens (approximate 10mm × 10mm of the enamel surface was exposed) per 147 treatment group were prepared to either: a) Polished surface group with 600-grit Silicon 148 Carbide grinding paper (SiC) ground following 3 µm diamond finish for enamel abrasion, b) 149 Partially roughened surface group with 400-grit SiC ground finish for surface polishing, or c) 150 Roughened surface group with 280-grit SiC ground finish for in vitro stain removal. 5 151 minutes ultrasonication in tap water was applied following each treatment to remove any 152 residual grinding/polishing materials. 153

154

155 *2.3. Tooth staining*

The tooth staining assay previously reported was used [27]. Freshly combined solutions 0.1%
(w/w) tannic acid (ACS reagent, Sigma-Aldrich) and (0.1% (w/w) of diammonium iron (II)
sulphate 6-hydrate (Sigma-Aldrich) form a dark colloidal iron (III) tannic acid complex
("ferric-tannate") on contact with air, which mimics a dietary tannin staining. The fresh

mixture was applied as successive layers on the enamel specimens for tooth staining, with the
initial layer, a 40 µl aliquot of the mixture was pipetted onto each specimen and dispersed
evenly over the specimen surface before air drying. For the subsequent 9 layers, 10 µl
aliquots of the solutions were applied as described above. Each layer was dried at 40 °C in an
oven (D-63450 Hanau, Kendro Laboratory Products Ltd, Germany) for 10 mins before
application of the subsequent layer.

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2.4. Toothbrushing protocol

168 The toothbrushing protocol used has previously been reported [27-29]. Eight bovine enamel specimens per treatment group were mounted in two brushing channels of an in vitro 169 brushing simulator and the experimental setup has been previously reported [29]. Oral B P35 170 medium toothbrushes were used for the brushing. A test band of the enamel specimen was 171 exposed when the un-brushed reference area was covered by ADA/ISO standard tape. 172 Slurries were generated with the addition of 1% (w/w) calcined alumina abrasives in 10% 173 (w/w) Glycerol (VWR International BVBA, Belgium) plus 0.5% (w/w) Hercules 7 MF 174 Carboxymethyl Cellulose (Hercules Incorporated, USA). A brushing load of 150 g was 175 applied and 150 g slurry was used in each channel [29]. Specimens were "brushed" at a 176 brushing speed of 120 rpm for up to 10,000 strokes and a temperature of 20°C was 177 maintained throughout the brushing procedure. 178

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2.5. Surface profiles of enamel specimens

Surface profiles were obtained before and after brushing using a Talysurf Series 2 inductive gauge profilometer (Taylor-Hobson, UK). Linear profiles (2D) were obtained on the surfaces with a point spacing of 0.25 µm and at a measurement speed of 0.5 mm/s. The wear depth and arithmetic mean surface roughness (Ra) values were calculated (µltra version 5.1.14,

185	Taylor-Hobson, UK). The inductive gauge profilometer uses a conical probe with 2 μ m				
186	diamond tip to accurately measure surfaces at the sub-micron level, has a resolution of 16 nm				
187	and a 1 mm range in the z-axis.				
188					
189	2.6. Gloss measurements				
190	A Novo-Curve small area glossmeter (Rhopoint Instruments Limited, UK) was used and				
191	gloss measurements were determined before and after brushing at intervals of 90 degrees				
192	rotation about the centre point of each specimen.				
193					
194	2.7. Colour evaluation				
195	Changes in colour were determined as previously described [27]. All surfaces were				
196	consistently dried prior to colour measurements. A calibrated spectrophotometer (Minolta				
197	CM-2600d, Konica Minolta Sensing Americas, Inc, USA) was used to measure colour values				
198	(L*, a*, b*) for each tooth specimen before staining (=Initial), after 10 layers of stain				
199	application (=Stained) and after the brushing treatments (=Brushed). The a* and b* values				
200	represent two colour axes, with a* the red-green axis and b* the yellow-blue axis. The L*				
201	value represents the value of 'brightness/darkness' of a colour, such that a perfect reflecting				
202	diffuser has an L* values of 100 and the perfect black body has an L* value of zero. The stain				
203	removal was assessed using the following formula:				
204	% $Removal = \frac{L^* (Brushed) - L^* (Stained)}{L^* (Initial) - L^* (Stained)} \times 100$				
205	Where L* (Initial), L* (Stained) and L* (Brushed) is the brightness before staining, after 10				
206	cycles of stain application and after toothbrushing for the requisite number of strokes with				
207	alumina abrasive slurry, respectively.				

Images of the enamel surfaces before staining, after 10 layers of stain and post-stain removal
with 1,000 brush strokes were digitally captured (Nikon D7000 camera, Nikon Corporation)
to visually demonstrate the effects of stain removal.

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2.8. Statistical analyses of the data

Single factor ANOVA was used for the data analyses with a significance level of $p \le 0.05$ applied. Pearson correlation coefficient was used to determine the linear relationship between two variables, and is denoted by r. It has a value between -1 and +1 inclusive, where -1 is perfect negative linear correlation, 0 is no linear correlation, and +1 is perfect positive linear correlation.

219

220 3. Results

3.1. Characterisation of calcined alumina abrasives

222 The particle size distributions of the alumina abrasives are shown in Figure 1 and Table 1.

223 The volume distribution is defined as the distribution per volume of the particle sizes, shown

as Volume %, a differential of total volume of all counts.

225 Representative SEM micrographs of the test calcined alumina abrasives are shown in Figure

1. Differences in particle size and range of morphologies for the four calcined alumina

227 abrasives can be clearly observed. All the abrasive particles were irregular shape and

consisted of a mixture of relatively small and large particles. SEM micrographs clearly

demonstrated that ultrafine is the finest, then followed by 3 μ m and 9 μ m, and 20 μ m is the

230 largest in terms of particle sizes. For the ultrafine abrasive, agglomerates were observed due

to its very fine particle size and individual particles were smaller than 1 μ m.

232

233 *3.2. Abrasivity of calcined alumina abrasives on polished enamel*

Table 2 provides wear depth and surface finishing of the polished enamel specimens after 234 brushing up to 10,000 strokes with the calcined alumina abrasives. Wear occurred for all the 235 tested enamel specimens, and the wear depth increased as the number of toothbrushing 236 strokes increased. A linear relationship ($r^2=0.99$) was identified between brushing strokes and 237 wear depth (enamel loss) for the tested alumina abrasives (Figure 2). Statistically significant 238 differences (p < 0.05) were detected in wear depth between the four calcined alumina 239 240 abrasives. The ultrafine caused the least wear (least abrasive), followed by the 9 µm and 20 μm abrasives (medium abrasive), while the 3 μm abrasive produced the most wear (most 241 242 abrasive).

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There was a decrease in gloss for all of the polished enamel surfaces after toothbrushing with all four calcined alumina abrasives. Statistically significant differences were detected for the decreases in gloss when the polished enamel specimens were brushed with the four alumina abrasives. There was a trend for the decrease in gloss to be greater when the polished enamel specimens were brushed with the larger size abrasive particles.

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The polished enamel surfaces became roughened with the toothbrushing and there was an 250 increase in the surface roughness for all of the polished enamel surfaces. The increase in 251 surface roughness of the polished enamel specimens was lowest for the ultrafine abrasive, 252 followed by the 3 µm and 20 µm particles and the 9 µm particles generated the greatest 253 increase in surface roughness. The loaded abrasive particles acted by generating grooves in 254 the polished enamel surfaces which resulted in a roughening of the polished enamel surfaces, 255 hence, resulting in the surface roughness increase. Abrasive particles with different particle 256 size and distribution impacted differently on the surface roughness increase, and no 257

statistically significant differences in roughness increase was detected between the 9 μm and
20 μm alumina abrasives.

260

Changes in gloss and roughness were not linearly related with regards to the number of brush
strokes although greater changes in gloss and roughness were found with an increase in
brushing strokes used.

264

265 3.3. Polishing effect of calcined alumina abrasives on partially roughened enamel 266 Polishing effect results for the alumina abrasives on the partially roughened enamel 267 specimens after brushing up to 10,000 strokes are shown in Table 3. There were no 268 appreciable differences in wear depth during the first 3000 brushstrokes, especially for the 269 least abrasive ultrafine particles. The ultrafine calcined alumina caused the least wear (least 270 abrasive), with a ranking of 9 μ m < 20 μ m < 3 μ m particles for the amount of wear produced. 271

There was an increase in gloss for all of the roughened enamel surfaces due to the wear to the 272 partially roughened enamel specimens, and these specimens also showed a decrease in 273 surface roughness. The wear depth, gloss increase, and roughness decrease were not linear in 274 relation to the number of brush strokes and the greatest changes were seen during the initial 275 1000 brushstrokes applied. The relationships between these parameters and the number of 276 277 brushstrokes showed differences amongst the different alumina abrasives. The most abrasive particles increased the gloss and decreased the surface roughness at a greater rate than the 278 least abrasive particles. 279

280

Notably, there was a continual increase in gloss and decrease in surface roughness for
roughened enamel surfaces brushed with the ultrafine particles throughout the entire brushing

protocol, up to 10,000 strokes. In contrast, for the 3 µm abrasive particles there was only a
marginal decrease in gloss and increase in surface roughness at 10,000 brush strokes
compared with the use of 5000 brush strokes.

286

287 Compared with the 20 µm abrasive, the other three abrasives (ultrafine, 3 µm and 9µm
288 particles) showed an enhanced performance on the finish of roughened enamel surfaces in
289 terms of gloss increase and surface roughness decrease.

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3.4. Stain removal on partially roughened enamel

The *in vitro* stain removal efficacy results are presented in Figure 3a. Data indicate that the 3 µm abrasive demonstrated the greatest cleaning power, followed by the 9 µm and 20 µm, and ultrafine abrasive which removed least stain. The images of enamel surfaces before staining and post-stain removal following application of 1000 brush strokes are shown in Figure 3b, relatively minimal stain was removed from the enamel surfaces after brushing when using the ultrafine abrasive. Increased stain was removed when brushing with the 20 µm and 9 µm abrasives, and the 3 µm abrasive removed the most stain from the stained surfaces.

299

300 4. Discussion

Both the individual particle sizes and the size distributions of the abrasive particles used in toothpastes are important factors in determining abrasivity and stain removal [12]. In the present study, the effect of calcined alumina abrasives with defined particle size and size distribution were investigated on toothbrush abrasion, polishing and *in vitro* stain removal. Results under these experimental conditions demonstrated that there exists a CPS for the tested calcined alumina abrasive particles. This is the first time that this has been

demonstrated with a tooth model system and data support the hypothesis underpinning thisstudy.

309

4.1. Critical particle size of the tested calcined alumina abrasives

311 *4.1.1. Critical particle size for abrasivity of polished enamel*

Size effect is a well-known phenomenon in abrasion with two-body wear a result of direct 312 313 contact and three-body wear due to surfaces being abraded by an "intervening slurry of abrasive particles" [30]. Furthermore, there is reportedly a minimum abrasive particle size or 314 315 CPS which allows maximum abrasive action [16,19,20]. The relationship between enamel abrasivity and particle size after 10,000 brush strokes are shown in Figure 4a. It is apparent 316 that there is a CPS of $\sim 2.3 \ \mu m$ for d10, 4.3 μm for d50 and 7.8 μm for d90 for the calcined 317 alumina abrasives. Notably, no CPS has previously been studied or reported in the literature. 318 A linear relationship was however found between particle size and enamel abrasion when 319 enamel was brushed with bioactive glass-based toothpastes (particle size range of 5-65 µm) 320 [23], calcium carbonate (median particle size range of 1-13 μ m) and dicalcium phosphate 321 (median particle size range of 1-16 µm) [21]. Larger particles were shown to have a higher 322 dentine abrasion when brushed with calcium carbonate and aluminium hydroxide. Similar 323 results were reported with smaller particles and narrow distributions of the particle size 324 reducing the magnitude of enamel wear when silica abrasives with a range of 4-12 µm 325 326 particle size were used. No impact of particle size on enamel erosion/abrasion was reported when eroded enamel was brushed with commercial toothpastes which contained abrasive 327 particles above 40 µm in size [25]. Notably, no clear association between average particle 328 size and average striation width (enamel loss) was shown when enamel was brushed with 329 commercial silicon carbide grit with a particle size range of 14-73 µm [26]. 330

331

4.1.2. Critical particle size effects on polishing and in vitro stain removal for partially
 roughened enamel

Unlike abrasivity and stain removal, a cleaning property of toothpaste that has received little

attention is its polishing ability [7]. High enamel lustre is important as more highly polished 335 enamel surfaces appear whiter than duller enamel surfaces and they do not accumulate 336 extrinsic stain. These properties are also important for the aesthetics of the dentition. 337 Furthermore and more importantly, smoothed and polished tooth surfaces are less receptive 338 339 to the build-up and retention of dental plaque. Indeed, the effects of enamel surface finish on 340 in vitro stain removal have confirmed that it requires fewer brush strokes to remove stain from polished compared with roughened enamel surfaces [27]. 341 342 Dulled enamel specimens have also been used to study polishing effects [7,8] and previous 343 work has used tooth specimens etched with acid prior to toothbrushing. In our study, 344 partially roughened enamel specimens were used to analyse the polishing effects and results 345 have now confirmed that a CPS also exists for the polishing effects of toothpaste abrasives 346 using this approach (see Figure 4b). Indeed, a CPS of ~2.3 µm for d10, 4.3 µm for d50 and 347 348 7.8 µm for d90 for the calcined alumina abrasives is apparent.

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334

Only one study has thus far investigated the potential for correlation between stain removal and particle size and no significant impact of particle size on tooth cleaning when brushing with perlite abrasive particles was detected [8]. However, we have now demonstrated CPS for *in vitro* stain removal of ~2.3 μ m for d10, 4.3 μ m for d50 and 7.8 μ m for d90 for the calcined alumina abrasives (see Figure 4c).

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356

4.2. Interrelationship between stain removal and tooth wear

Both the individual particle sizes and size distributions of the abrasive particles are important 357 factors in determining abrasivity and stain removal. It is therefore not surprising that 358 abrasivity and stain removal are correlated [12]. Normally more stain is removed when 359 brushed with more abrasive particles. In the present study the 3 µm particles are the most 360 abrasive, therefore it has greatest cleaning power and the maximum stain removal is at 3 µm. 361 A linear relationship was detected under the present experimental conditions between enamel 362 363 abrasivity (at 10,000 brush strokes) and *in vitro* stain removal efficacy (1000 brush strokes) (data not shown). Similar results have been reported elsewhere indicating the relationship 364 365 between dentine abrasivity and stain removal [31,32]. Interestingly however exceptions to this have also been reported, and some data show that for a toothpaste with improved 366 cleaning, increased dentine abrasivity is not a pre-requisite. This can potentially be explained 367 due to the different influence on dentine and stains by factors such as abrasive type, particle 368 surface and size, as well as the chemical influence of other toothpaste components [33]. 369

370

371 *4.3. Finishing and polishing mechanism*

Results for the roughened enamel surfaces showed that brushing with alumina abrasives 372 produced wear, increased gloss and decreased surface roughness. Gloss increased and surface 373 roughness decreased with the increasing number of brushstrokes. Notably, there was a peak 374 at which the increase in gloss and decrease in surface roughness was optimal. After that peak, 375 376 further brushing resulted in the surface becoming rougher and exhibited a lower gloss finish. Interestingly, the number of brushstrokes required to reach the optimal peak of surface finish 377 was different for the various abrasives used. When the partially roughened enamel surfaces 378 were brushed with abrasive particles, there was removal of asperities through wear (abrasion) 379 which reduced the size of the peaks and troughs and therefore smoothed the partially 380 roughened surfaces thereby decreasing surface roughness. Different abrasive treatments 381

impact the removal of asperities at different rates, therefore requiring different brushstrokes
to reach the optimal peak of surface finish. Having more abrasive particles results in a
requirement for a fewer number of brushstrokes.

385

386 This phenomenon may be explained by the stages described below.

387 Stage 1: There is a partially roughened enamel surface prior to the commencement of388 brushing.

Stage 2: The partially roughened surface would subsequently become smoother due to the increasing number of brushstrokes allowing for some of the peaks and troughs previously present on the roughened surface to be removed due to abrasion. Consequently, the gloss would increase and the surface roughness would decrease.

Stage 3: The finish and polish would reach a maximum effect with further brushing, and
surface peaks and troughs would be removed resulting in a smoother surface. The gloss
would therefore achieve the highest level and surface roughness would be at its lowest level.
Stage 4: The smooth surface would become gradually roughened due to further brushing.

397 Consequently, compared with stage 3, the gloss would decrease, surface roughness would

398 increase, as would wear depth.

399

Our data indicate that there are differences in the number of brushstrokes to reach each of the stages described above for the different abrasives used. Generally, the most abrasive particles would require fewer brushstrokes compared with the least abrasive particles, to reach each stage. Indeed, it was notable that there was a continual increase in gloss and decrease in surface roughness for partially roughened enamel surfaces brushed with the ultrafine calcined alumina particles throughout the whole brushing procedure up to 10,000 strokes. In contrast,

for the 3 μm abrasive particles there was a marginal decrease in gloss and increase in surface
roughness at 10,000 brushstrokes compared with data from brushing at 5,000 strokes.

408

409 5. Conclusions

Results from the present study have confirmed the existence of a critical particle size (CPS) 410 for the first time in a tooth model system and a CPS of ~2.3 µm for d10, 4.3 µm for d50 or 411 7.8 µm for d90 for the calcined alumina abrasives was apparent. Abrasive particle size 412 413 affected the wear of the polished enamel surfaces, the finish of the partially roughened enamel surfaces and the in vitro stain removal on the roughened enamel surfaces. This 414 dimension identified enabled maximum abrasive action on the tested specimens resulting in 415 the largest wear depth, greatest surface polishing and best stain removal. The novel findings 416 provide new information on abrasive particle size for modification and control of toothpaste 417 abrasivity and cleaning, supporting the development of new toothpaste formulations which 418 can harness optimal abrasive particle size and size distributions for tooth cleaning. 419

420

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449 Table 1 Particle size characteristics of the tested calcined alumina abrasives*

Abrasive	d10 [µm]	d50 [µm]	d90[µm]
Calcined Alumina ultrafine	1.1	2.2	5.2
Calcined Alumina 3 µm	2.3	4.3	7.8
Calcined Alumina 9 µm	4.4	7.4	11.9
Calcined Alumina 20 µm	8.2	14.0	23.2

451 * Data are average values of three measurements. The d10, d50 and d90 values indicate that

10%, 50% and 90% of the particles measured were less than or equal to the size stated.

463	Table 2	Surface finish and wear depth of polished bovine enamel ($n=8$, mean \pm standard deviation)
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Brush strokes	Parameters	ultrafine	3 μm	9 μm	20 µm
Dofono	Gloss, GU	107.3±1.1	108.1±1.0	$108.4{\pm}1.1$	108.7±0.7
Delore	Roughness, µm	0.036 ± 0.009	0.037 ± 0.004	0.031 ± 0.005	$0.034{\pm}0.005$
	Gloss, GU	101.2±3.2	89.7±3.2	81.0±6.8	73.5±5.0
	Gloss change, GU	-6.1±2.8	-18.4±2.8	-27.4±5.9	-35.3±4.8
1000	Roughness, µm	0.042 ± 0.013	0.048 ± 0.009	0.060 ± 0.007	$0.055 {\pm} 0.006$
	Roughness change, µm	0.006 ± 0.015	0.012 ± 0.008	0.029 ± 0.008	0.021 ± 0.010
	Wear depth, µm	0.058 ± 0.013	0.182 ± 0.109	0.107 ± 0.034	$0.156{\pm}0.075$
	Gloss, GU	100.9±4.1	85.6±4.2	74.3±8.3	65.7±4.2
	Gloss change, GU	-6.4±4.1	-22.5 ± 3.6	-34.1±7.4	-43.0±4.1
3000	Roughness, µm	0.051 ± 0.016	0.057 ± 0.007	0.075 ± 0.018	0.072 ± 0.014
	Roughness change, µm	0.015 ± 0.019	0.020 ± 0.008	0.044 ± 0.020	$0.039{\pm}0.013$
	Wear depth, µm	0.094 ± 0.025	0.393 ± 0.142	0.262 ± 0.100	0.276 ± 0.094
	Gloss, GU	101.0±4.4	83.4±3.6	73.5±7.7	64.5±6.3
	Gloss change, GU	-6.3±4.1	-24.7±3.3	-34.9±6.9	-44.2 ± 6.0
5000	Roughness, µm	0.043 ± 0.006	0.065 ± 0.008	0.085 ± 0.018	$0.082{\pm}0.011$
	Roughness change, µm	0.007 ± 0.012	0.028 ± 0.007	0.053 ± 0.018	0.049 ± 0.012
	Wear depth, µm	0.110 ± 0.030	0.675 ± 0.226	0.471 ± 0.202	0.421 ± 0.109
	Gloss, GU	102.5 ± 4.9	78.3±7.1	69.2±9.6	60.2 ± 6.6
	Gloss change, GU	-4.8±4.4	-29.8±6.5	-39.2±8.8	-48.6±6.4
10000	Roughness, µm	0.054 ± 0.011	0.099 ± 0.023	0.110±0.033	0.105 ± 0.020
	Roughness change, µm	0.017 ± 0.015	0.063 ± 0.023	0.079 ± 0.032	0.071 ± 0.022
	Wear depth, µm	0.204 ± 0.070	1.376 ± 0.439	0.833 ± 0.372	$0.778 {\pm} 0.200$

468	Table 3	Surface finish and wear depth of partially roughened bovine enamel ($n=8$, mean \pm standard deviation)
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Brush strokes	Parameters	ultrafine	3 μm	9 μm	20 µm
Dafara	Gloss, GU	17.6±5.8	22.0±6.8	17.3±6.0	$18.0{\pm}4.9$
Delore	Roughness, µm	0.151±0.023	0.124 ± 0.036	$0.146{\pm}0.016$	0.146 ± 0.027
	Gloss, GU	46.2±6.9	76.8±10.7	62.3±6.7	47.9±6.5
	Gloss change, GU	28.6 ± 3.6	54.7±6.1	45.1±6.0	30.0±3.1
1000	Roughness, µm	0.109 ± 0.015	$0.078 {\pm} 0.020$	$0.094{\pm}0.014$	0.116 ± 0.018
	Roughness change, µm	-0.043 ± 0.011	-0.046 ± 0.024	-0.052 ± 0.016	-0.031±0.013
	Wear depth, µm	0.216 ± 0.026	0.216 ± 0.062	$0.195{\pm}0.021$	$0.317{\pm}0.088$
	Gloss, GU	55.6±2.4	$84.6 {\pm} 7.8$	73.6±5.0	57.2±5.2
	Gloss change, GU	38.0 ± 5.5	$62.6{\pm}6.7$	56.4 ± 4.8	39.2±4.1
3000	Roughness, µm	0.0928 ± 0.0110	0.0697 ± 0.0167	0.0828 ± 0.0121	0.1111 ± 0.0210
	Roughness change, µm	-0.0582 ± 0.0155	-0.0539 ± 0.0270	-0.0629 ± 0.0107	-0.0351±0.0263
	Wear depth, µm	0.121 ± 0.0222	0.3694 ± 0.1684	0.2405 ± 0.0566	0.4256 ± 0.1380
	Gloss, GU	$65.8 {\pm} 4.0$	84.9±6.3	76.1±3.3	60.4 ± 4.9
	Gloss change, GU	48.2 ± 8.2	62.9 ± 7.9	58.8±4.6	42.4±5.5
5000	Roughness, µm	0.083 ± 0.008	0.071 ± 0.015	$0.082{\pm}0.008$	$0.104{\pm}0.012$
	Roughness change, µm	-0.068 ± 0.019	-0.053 ± 0.031	-0.064 ± 0.017	-0.043 ± 0.022
	Wear depth, µm	0.213±0.038	0.619 ± 0.257	$0.380{\pm}0.147$	$0.492{\pm}0.186$
	Gloss, GU	90.2±6.3	79.9±8.3	73.4±4.1	60.9 ± 6.0
	Gloss change, GU	72.6±9.2	57.9±8.2	56.1±7.7	42.9±6.5
10000	Roughness, µm	0.065 ± 0.005	0.102 ± 0.027	$0.098{\pm}0.008$	$0.108{\pm}0.019$
	Roughness change, µm	-0.086 ± 0.023	-0.021 ± 0.040	-0.048 ± 0.019	-0.038 ± 0.023
	Wear depth, µm	0.223 ± 0.050	1.226 ± 0.584	$0.707 {\pm} 0.401$	0.696±0.312

473 *Figure legends*

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Figure 1 Characteristics of the tested calcined alumina abrasives.

Particle size distribution: Particle size distributions of the calcined alumina abrasives determined by using Malvern Mastersizer 2000 laser diffraction particle size analyser. Data are average values from three measurements. The volume distribution is defined as the distribution per volume of the particle sizes, shown as Volume %, a differential of total volume of all counts.

SEM observation: Representative SEM micrograph of the tested calcined alumina abrasives particles. a) ultrafine; b) 3 μ m; c) 9 μ m; and d) 20 μ m. Differences in particle size and range of morphologies for the four calcined alumina abrasives can be clearly observed. For the ultrafine abrasive, agglomerates were observed due to its very fine particle size and individual particles were smaller than 1 μ m. All the abrasives consisted of a mixture of relatively small and large particles

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Figure 2 Relationship between brush strokes and enamel loss (wear depth) obtained from polished bovine enamel up to 10,000 brush strokes for the abrasive particles studied. A linear relationship ($r^2=0.99$) was found between brushing strokes and wear depth (enamel loss, shown as mean and standard deviation) for the tested alumina abrasives.

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Figure 3 a) Stain removal efficacy for roughened bovine enamel specimens after 1000 strokes when brushed with calcined alumina abrasives; b) Representative images of enamel surfaces before staining, after staining and post-stain removal with 1000 brush strokes. (left to right): before stain; after staining; stain brushing with ultrafine abrasive; stain brushing with 3 μ m abrasive; stain brushing with 9 μ m abrasive; stain brushing with 20 μ m abrasive.

Figure 4 The relationship between enamel abrasivity, polishing capability (gloss change), and *in vitro* stain removal efficacy against abrasive particle size (d10, d50 and d90). The d10, d50 and d90 values indicate that 10%, 50% and 90% of the particles measured were less than or equal to the size stated.

- a) Enamel wear depth of polished specimens after 10,000 brush strokes plotted against calcined alumina particle size (d10, d50 and d90);
- b) Gloss change of partially roughened enamel specimen surfaces after 3,000 brush strokes plotted against calcined alumina particle size (d10, d50 and d90);
- c) Stain removal efficacy of roughened enamel specimens after 1,000 brush strokes plotted against calcined alumina particle size (d10, d50 and d90).



505 Figure 1







